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Growth and Characterization of GaAsP Nanowires on GaAs(111)B Substrate by Selective-Area Metal Organic Vapor Phase Epitaxy

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1. Introduction

Over the past several years, semiconductor nanowires (NWs) have attracted a great deal of attention because they might become basic active elements in next-generation integrated circuits and functional devices [1]. Field-effect transistors [2], single-electron transistors [3], light-emitting devices [4], and chemical sensors [5] using NWs have been fabricated to demonstrate their feasibility and performance. These NW devices have also become the focus of great interest because they could bring us new physics on quantum-size effects by electrons and holes contained in one-dimensional space. To make a NW device and observe quantum-size effects, it is crucial to control the size and position of NWs on a substrate surface and to establish the method of fabricating vertical heterostructure NWs. We have reported on the growth of III-V semiconductor NWs by selective-area metal organic vapor phase epitaxy (SA-MOVPE) and have succeeded in fabricating InGaAs/GaAs vertical heterostructure NWs [6]. Moreover, we have attempted to fabricate AlGaAs/GaAs vertical heterostructure NWs, which would be another candidate that can extend the range of materials for device applications. However, it has been difficult to suppress the lateral growth of AlGaAs. Because the growth temperature (800°C) of AlGaAs NWs is about 50°C higher than the optimum growth temperature of GaAs NWs, the grown shape of GaAs on top of AlGaAs NWs has not been a pillar but nearly planar at temperatures of 800°C and higher. Thus, it has been a challenge to form vertical heterostructures. Here, we have focused on GaAsP NWs that are expected to suppress laterral growth, thereby enabling us to form GaAsP/GaAs vertical heterostructure NWs. We will explain the growth process of GaAsP NWs obtained by SA-MOVPE and the grown structures characterized by scanning electron microscopy (SEM). We will then discuss the optical characterization of GaAsP NWs by measuring micro-photoluminescence (μ-PL).

2. Experimental Procedures

The fabrication process for GaAsP NWs (Fig. 1) started with the preparation of patterned GaAs(111)B substrates partially covered with an SiO_2 mask for SA-MOVPE. After a 25-nm-thick SiO_2 film was deposited on the GaAs(111)B substrate by plasma sputtering, hexagonal-opening patterns were defined using electron-beam (EB) lithography and wet-chemical etching based on buffered hydrofluoric acid (BHF). The SiO_2 patterns were designed to have a periodic array of openings with a diameter from 50 to 250 nm and a pitch from 0.5 to 3.0 μm. The SA-MOVPE growth of GaAsP was carried out in a horizontal MOVPE system working at a pressure of 0.1 atm. The source materials were trimethylgallium (TMG), tertiarybutylphosphine (TBP), and arsine (AsH_3) (20% in hydrogen). The growth temperatures ranged between 750 and 775°C and the growth time was 20 min. The partial pressures of TMG were from 2.7×10^{-6} to 4.1×10^{-6} atm, those of TBP were from 4.2×10^{-5} to 2.5×10^{-4} atm, and those of AsH_3 were from 4.2×10^{-5} to 2.5×10^{-4} atm. The (TBP+AsH_3)/(TMG) ratios (V/III ratios) were from 20 to 185.

The μ-PL was measured at 4.2 K. Excitation light from a He-Ne laser (wavelength: λ = 632.8 nm) was focused on the NW arrays using ×50 microscope objectives with 0.42-N.A., which were also used to collect the PL from NWs. The excitation power was 1.0 kW/cm^2, and the laser spot was less than 2 μm in diameter.

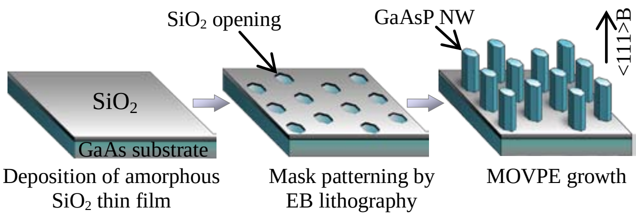

Fig. 1: Schematic of nanowire fabrication process.

3. Results and Discussion

We investigated how the shape of grown GaAsP NWs changed when we altered the V/III ratios under a constant growth temperature, partial pressure of TMG, and growth time. Figs. 2(a) to (c) are SEM images of GaAsP NWs grown at 750°C with V/III ratios (TMG supply was constant and (TBP+AsH_3) supply was changed) corresponding to 185, 62, and 31. The pattern pitch and mask opening diameter are 1 μm and 100 nm. For the GaAsP NWs grown at the V/III ratio of 185 (Fig. 2(a)), we can see a large amount of planar deposition on the patterned area, which was due to the lateral growth of GaAsP. The degree of planar deposition was highest at the V/III ratio of 185, but the deposition decreased in size as the V/III ratio was decreased and its extent was lowest at the V/III ratio of 31 (Figs. 2(b) and (c)). We thought the cause of the dependence of deposition on the V/III ratio was related to the concentration of P atoms on the substrate surface. When the

V/III ratio was higher than 62, i.e., the concentration of P atoms was in a larger range, the adsorbed P atoms prevented Ga and As atoms from adsorbing onto the (111)B growth interface, resulting in planar deposition with a low density of NWs grown in between the depositions. The NW height became maximum at the V/III ratio of 62 but it decreased as the V/III ratio was decreased from 62 to 31. We think this was due to a decreased supply of group-V sources.

Next, we tried another condition under which to grow GaAsP NWs with uniform hexagonal structures without planar deposition. We found that the condition for reduced lateral growth of GaAsP NWs was the V/III ratio of 31, but the NW height was too short. To solve this issue, we increased the partial pressure of TMG because the growth rate in the axial direction was dependent on the strength of the group-III source. Fig. 3 has SEM images of GaAsP NWs grown at a V/III ratio of 20, the condition for the increased TMG supply. We can see the NWs have hexagonal and triangular structures and crystal facets, which indicates the formation of {-110} equivalent side facets and a top (111)B facet.

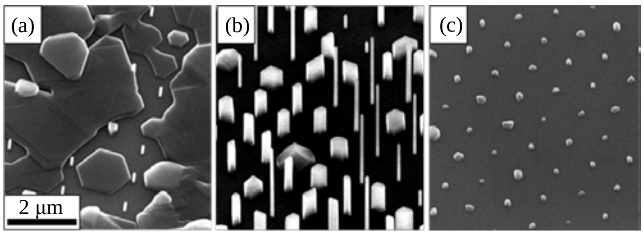

Fig. 2: SEM images of GaAsP NWs grown at 750°C. V/III ratios of GaAsP NWs are (a) 185, (b) 62, and (c) 31.

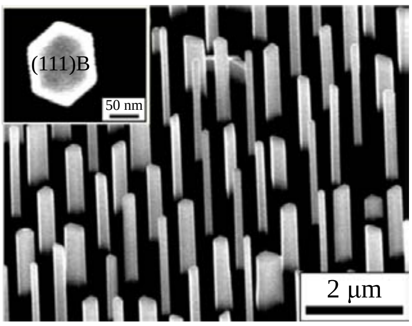

Fig. 3: SEM images of GaAsP NWs grown at 750°C with V/III ratio of 20.

Fig. 4 shows the PL spectrum of GaAsP NWs grown at the V/III ratio of 20. The PL spectrum of the GaAs substrate has also been plotted along the blue line for reference. The number of NWs excited by the laser beam was about 3-5 for the sample with the pitch of 1 µm. The PL spectrum of GaAsP NWs exhibited a strong emission peak at a photon energy of 1.79 eV, which is clearly different from the main peak of the GaAs substrate. The full width at half maximum (FWHM) of the PL spectrum of the NWs was 18 meV. The PL peak energy and the FWHM did not change when we changed the position of laser excitation on the sample surface. The FWHM of the GaAsP NWs was as narrow as those of the InGaAs NWs that have been reported [7]. This indicates that GaAsP NWs with an equal atomic composition were formed. We estimated the solid P atomic composition at 13% from the PL peak energy, i.e., $GaAs_{0.75}P_{0.25}$.

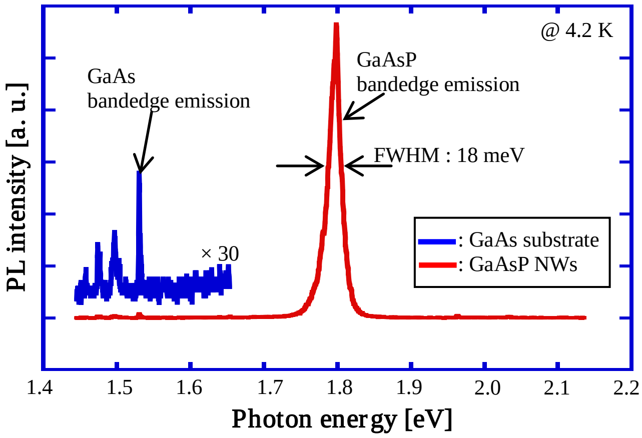

Fig. 4: PL spectra for GaAsP NWs and GaAs substrate at 4.2 K.

4. Conclusions

We fabricated GaAsP NWs on GaAs (111)B substrates by using SA-MOVPE and analyzed the dependence of the shape of grown GaAsP NWs on the V/III ratio. We also conducted µ-PL measurements of GaAsP NWs at 4.2 K, and estimated the alloy composition from the position of the spectral peak. The results indicate that high-quality $GaAs_{0.75}P_{0.25}$ NWs were grown, which shows promise for fabricating vertical heterostructure NWs.

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