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Control of the size and the emission wavelength in InP-based nanowire quantum dots

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Abstract

We report on the control of lateral size of InP-based nanowires (NWs) by *in situ* annealing. InP NW with lateral size of 20 nm was obtained under optimized annealing conditions. The emission wavelength of InP/InAsP heterostructure NWs was controlled by controlling the supply ratio of group V source materials during the growth. By combining the size and wavelength control, we have obtained emission in the telecom band from quantum dots formed in NWs.

1. Introduction

Nanowire quantum dots (NW-QDs) are one of the promising candidates for single photon sources and InP-based NW-QDs are capable of operation in telecommunication band. Emission in the telecom bands from InP-based heterostructure NWs including NW-QDs have already been demonstrated by several groups including ourselves [1-4]. Further refinement of the reproducibility and purity of the single photons are required for quantum information processing and quantum cryptography applications. The size and shape of NWs greatly influences their electronic and optical properties such as quantum confinement [5], light extraction efficiency [6], and so on. In the case of InP-based NWs, it is necessary to make the diameters less than 20 nm for good lateral quantum confinement. Attempts to reduce the size of NWs by thermal etching has been attempted for GaAs-based NWs [5], but it is more challenging in InP-based NWs due to their lower dissociation temperature and higher dissociation pressure. Hence, in this study, we investigated the *in situ* thermal annealing of InP NWs in growth system towards the control of their size and shape. We also attempted the emission wavelength control of NW-QDs by controlling the alloy composition of InAsP.

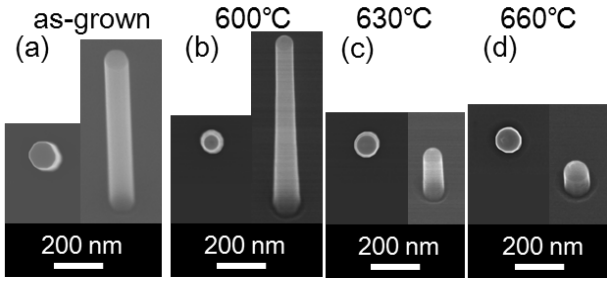

Fig.1: SEM images of InP NWs (a) prior to annealing, and (b)-(d) after annealing at each temperature for 20 min.

2. Experimental procedures

NWs were grown by metal-organic vapor-phase epitaxy (SA-MOVPE) on partially masked InP (111)A substrates. The mask pattern consisted of a periodic array of circular openings with diameter d_0 and arranged in the triangular lattice with pitch a. The source materials for the growth were trimethyindium (TMIn), tertiarybutylphosphine (TBP), and arsine (AsH_3). For InP NWs, the growth temperature and V/III ratio were set at 660 °C and 4.8, respectively. Annealing of InP NWs was carried out in the MOVPE reactor while feeding TBP with TBP partial pressure [TBP] of 3.24×10^{-4} atm. The annealing temperature T_A was varied from 600 to 660 °C, and the annealing time t_A was changed from 5 to 20 min. InAsP and InP were successively grown on InP NWs with and without annealing to form heterostructure NWs. To control the alloy composition y in $InAs_yP_{1-y}$, ratio of the partial pressure p_{As}, defined as $p_{As} = [AsH_3]/([AsH_3] + [TBP])$ was changed from 4 to 54 % during the growth of InAsP,

3. Results and discussions

Figure 1(a) shows InP nanowires prior to the annealing, and Figs. 1(b)(c) and (d) show the NWs after the annealing for 20 min. The mask opening diameter d_0 was 110 nm. Before annealing, the nanowires have hexagonal prismatic structures with vertical facets and an average diameter of about 120 nm. For T_A=600 °C, the nanowires became rounded and tapered,

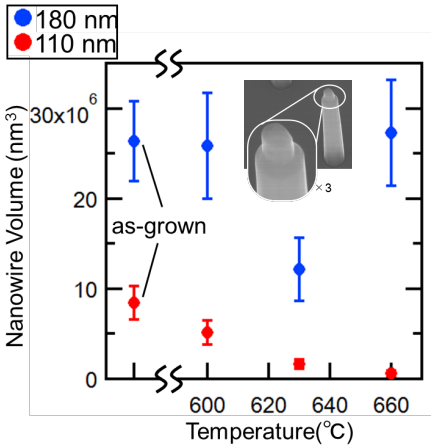

Fig. 2: Dependence of NW volume on the annealing temperature T_A and initial NW diameter d. Inset shows an SEM imager of an InP NW after annealing with T_A=660 °C with d=180 nm. Annealing time was 20 min.

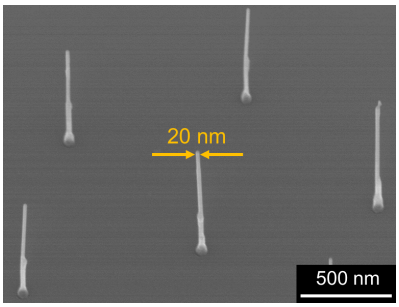

Fig. 3: SEM image of InP NWs after annealing under optimized conditions.

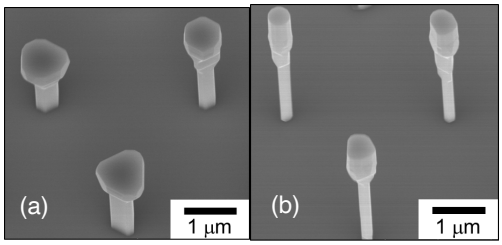

Fig. 4: SEM image of InP/InAsP/InP heterstructure NWs formed (a) without and (a) with annealing of the first InP NW.

and the sidewall facets were disappeared. As a result, the diameter of NWs was reduced to about 75 nm. In these results, the NW height was longer than that before annealing, indicating that the mass transport took place. On the other hand, the height of the nanowires was found to decrease drastically after the annealing at 630 and 660 °C. This is due to the desorption of In and P atoms at these temperatures, and it is not suitable for the size control under these conditions.

It was found that the desorption and mass transport of In were dependent on the initial diameter of NWs. Figure 2 shows the annealing temperature dependence of the volume of NWs with diameter d of 110 nm and 180 nm. As mentioned above, In desorption was confirmed in NWs with d=110 nm, while the volume was almost independent of the annealing temperature for NWs with d=180 nm. We also found that, at T_A=630 and 660 °C with d=180 nm, much thinner and short segments of NWs were formed at the top, as typically shown in the inset of Fig. 2. We speculate these thin and short segments were formed via self-catalyzed vapor-liquid-solid (VLS) growth mechanism, implying that In droplets were formed on the top of the NWs during annealing, followed by the mass transport of In. This VLS mechanism was found to be effective to realize thin NWs. Based on these results, we obtained InP NWs with diameter of 20 nm with T_A=600 °C and t_A=5 min, with d_0=60nm, as shown in Fig. 3.

Figure 4 shows SEM images of InP/InAsP/InP heterostructure NWs. InAsP layer was grown with and without annealing of InP NWs for Fig. 4(a) and (b), respectively. Formation of NWs with lar larger top was commonly observed in our InP/InAsP heterostructure NWs [7]. It is apparent that diameter of NWs formed after annealing is much smaller than those formed without annealing. Detail comparison of the size of two types of NWs are summarized in Table I. Note that the NWs formed with annealing showed bimodal height distribution, which is thought to be originated from the size

Table 1: Comparison of the size of heterostructure NWs formed with and without annealing of InP prior to the growth of InAsP.

	Bottom diameter (nm)	Top diameter (nm)	height (nm)	
without annealing	440 ± 90	670 ± 130	1750 ± 450	
with annealing	190 ± 50	470 ± 70	3200 ± 210	4260 ± 200

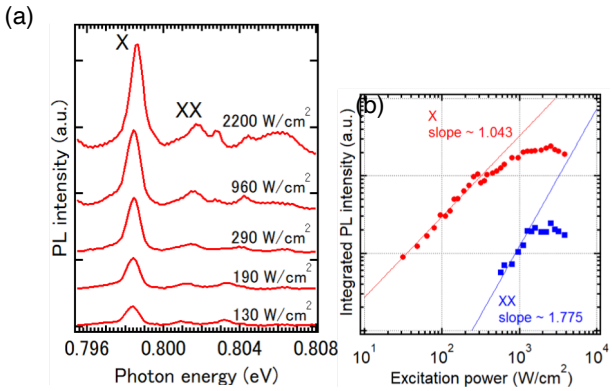

Fig. 5: (a) PL spectra and its excitation intensity dependence of InP/InAsP heterostructure NWs. (b) Integrated intensity vs excitation intensity of exciton (X) and biexciton (XX)

reduction via VLS mechanism or conventional mass transport.

The emission wavelength from InAsP can be controlled by controlling the supply ratio p_{As} of AsH_3. In the case of samples formed without annealing, the emission wavelength of InAsP QDs in InP NWs were systematically red-shifted from 1.216 to 1.473 μm by increasing p_{As} from 4 to 32 % (not shown here). Figure 5(a) shows photoluminescence spectra of heterostructure NWs formed with annealing and p_{As}=54%. A sharp peak with full width of half maximum (FWHM) of 770 μeV was observed at 1.553 μm, which was attributable to InAsP QDs formed in NWs [7]. It was found that the average FWHM of the NW-QDs formed with annealing was 720 μeV, and was narrower than the samples without annealing (940 μeV), This is thought to be due to the stronger lateral confinement owing to the miniaturization in annealed NWs and to the suppression of the spectral diffusion originating from the Stark effect.

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