# Single Photon Emission from InAsP Quantum Dots Embedded in Density-controlled InP Nanowires

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## Abstract

We attempted the density control of InP-based nanowires (NWs) and nanowire quantum dots (NW-QDs) in selective-area metalorganic vapor phase epitaxy. InP nanowire arrays in 5  $\mu$ m pitch with average diameter of 67 nm was successfully grown via optimization of growth conditions. InAsP quantum dots were embedded in those density-controlled InP NW arrays, and clear single photon emission and behavior of exciton-biexciton cascaded emission was observed, showing a nature of single QDs.

## 1. Introduction

InP-based nanowires (NWs) are promising for photonic devices, because their band gap energy is compatible with optical fiber telecommunication. Combination of NWs and quantum dots (QDs) by embedding QDs inside NWs, or NW-QDs, is particularly promising for single photon sources used in quantum information and quantum cryptography application. Formation of NW-QDs using InP-based materials has been reported [1-3] towards a single photon emitter in the optical communication band, and single photon emission[2-3] and entangled photon emission [4] has been demonstrated. One of the advantages of NW-QD is that QDs are formed at predetermined positions by combination of lithography and epitaxial growth. Thus, if NWs are sufficiently apart each other, one can easily realize a single photon emitter without post-growth selection. In reality, however, the distance between NWs critically influences the dynamics of their growth, for instance, as reported in Ref. [5], and realizing low-density NWs with small diameter is not straightforward in selective-area metalorganic vapor-phase epitaxial (SA-MOVPE) growth [6].

In this report, we attempted the density control of InP NWs in SA-MOVPE. InP NW arrays with 5  $\mu$ m pitch was successfully formed. A single InAsP QD was also realized in the low-density InP NW array, showing clear photon antibunching in their excitonic photoluminescence.

#### 2. Experimental Procedure

Selective-area growth of InP-based NWs was carried out by MOVPE on InP (111)B substrate, on which periodic array of circular mask holes with diameter  $d_0$  were defined. The mask holes were arranged in a triangular lattice with a period *a*, and *a* was ranged from 1 to 6  $\mu$ m. Trimethylindium (TMIn), tertiarybutylphosphine (TBP), and arsine (AsH<sub>3</sub>) were used as source materials. In the present experiment, partial pressure [TBP] of TBP was fixed at  $1.05 \times 10^{-6}$ atm, but that of TMIn ([TMIn]) and growth temperature  $T_{\rm S}$ was varied. For characterization, scanning electron microscopy (SEM) and low-temperature photoluminescence (PL) measurement were used. Photon statistics was investigated by standard Hanbury-Brown Twiss measurement using superconducting-nanowire single photon detectors.

## 2. Results and Discussions

We first attempted the optimization of InP NWs with large a. Figure 1 summarizes results of InP-NWs growth and its dependence on the period of NW array. Here,  $d_0$  was 100 nm, and growth conditions were  $T_s$  of 605 °C and V/III ratio of 20, which was based on our standard growth condition for InP NWs [7,8]. When a was 1  $\mu$ m, uniform array of NWs were formed, as shown in Fig. 2(a). However, some selectively-grown InP showed anomalously thick shape when a was 5  $\mu$ m (Fig. 1(b)), and the uniformity was deteriorated as a becomes wider. This is summarized in Fig. 1(c), in which the formation rate of the NWs and their average diameter are plotted as a function of a. We considered that this deterioration was mainly due to the excess supply rate of In, because the In supply is determined by the surface and gas phase diffusion, and those for a single NW increases with a.

Based on this consideration, we decreased [TMIn], and the uniformity was improved as expected. Further im-



Fig. 1 SEM image of InP NWs with (a)  $a=1 \mu m$  and (b)  $a=5 \mu m$ . (c) Average diameter d of NWs and their formation rate.



Fig. 2 SEM images of InP/InAsP/InP NW-QD array with period  $a=5 \ \mu$ m.

provement of the uniformity was achieved by reducing  $d_0$ . For  $d_0 = 50$  nm and V/III ratio of 100, we obtained InP NWs with average diameter of 67 nm.

Next, InP NWs containing InAsP QD was grown, and the result is shown in Fig. 2. InAsP was grown at 580 °C for 3 sec with  $[TBP]=1.05\times10^{-4}$  atm, and  $[AsH_3]=5.25\times10^{-6}$  atm. Very thin InP NWs containing InAsP QDs were obtained as expected.

Figure 3 summarizes results of low-temperature PL measurement for a single NW-QD and its dependence on excitation intensity I. Two peaks, named as X and XX, were observed in the PL spectra, and their intensity linearly or superlinearly with I. These PL are thought to be an exciton and a biexciton in a single QD. It is noted that the peak XX appears at higher energy side than X. This antibinding of biexciton indicates that the electrons and holes are confined in the very small regions and suggests the formation of a small QD in a NW.

Furthermore, we studied the photon statistics of the emission from a NW-QD. Figures 4(a)-(c) show results of auto-correlation (X-X) and cross-correlation (X-XX) measurements under barrier excitation. Photon antibunching behavior in the X-X correlation were observed under pulsed and cw-excitation. The cw-like antibunching in the pulse-based correlation is caused by the carrier injection from InP NW over timescale of the X-decay of 4.72 ns, which was deduced by X-pulsed excitation laser correlation (not shown). Figure 4(c) shows a cascaded emission of an



Fig 3. PL spectra (a) and its excitation intensity dependence (b) of a single NW-QD.



Fig. 4 Results of photon-correlation measurement on a NW-QD. Inset Thick solid lines show results of fitting by using three level model (inset).  $\tau_{\rm X}$ =4.72 ns and  $\tau_{\rm XX}$ =3.67 ns are deduced by photon counting method. *G*= 0.20 ± 0.01,  $g_{\rm X}^{(2)}(0)$ = 0.19 ± 0.04, and  $g_{\rm XX}^{(2)}(0)$ = 0.79 ± 0.20 are fitting parameters.

exciton and a biexciton. Thick solid lines in Figs. 4(b)-(c) show results of fitting for each data by using a three level model (inset), which takes into account multi-photon generation MP1= $g_X^{(2)}(0)n_X(\infty)$  and MP2=  $g_{XX}^{(2)}(0)n_{XX}(\infty)$ . The experiments were nicely reproduced by the fitting and obtained  $g_X^{(2)}(0)$  values for two-fitting are consistent within experimental and fitting errors and was 0.19 ± 0.04.

# 4. Conclusions

We have succeeded in the density control of InP-based NWs by manipulating growth conditions in SA-MOVPE. NW-QDs have also been successfully formed in those density-controlled NWs, and their PL and photon statistics have shown clear nature of QDs.

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