Mater. Res. Soc. Symp. Proc. Vol. 1439 © 2012 Materials Research Society
DOI: 10.1557/opl.2012.912

Formation of a single In(Ga)As/GaAs quantum dot embedded in a site-controlled GaAs nanowire by metalorganic chemical vapor deposition for application to single photon sources

J. Tatebayashi[1], Y. Ota[1], D. Karunathillake,[1,2] S. Ishida[1,2], M. Nishioka[1,2], S. Iwamoto[1,2] and Y. Arakawa[1,2]
[1]*NanoQUINE, Univ. of Tokyo, 4-6-1, Komaba, Meguro-ku, Tokyo, 153-8505, JAPAN*
[2]*Institute of Industrial Science, Univ. of Tokyo, 4-6-1, Komaba, Meguro-ku, Tokyo, 153-8505, JAPAN*

ABSTRACT

We report the formation and optical properties of site-controlled InAs/GaAs quantum dots (QDs) embedded in GaAs nanowires (NWs) by selective metalorganic chemical vapor deposition for application to single photon sources. InAs/GaAs QD-in-NWs with various InAs thicknesses are realized on patterned GaAs(111)B substrates in the form of InAs/GaAs heterostructures and identified by structural analyses using scanning transmission electron microscopy and photoluminescence characterization. Sharp excitonic emission peaks at 10 K from single QD-in-NWs with the narrowest exciton linewidth of 87 μeV are observed. Light emission from the single QD-in-NW shows photon antibunching which evidences single photon emission from high-quality QD-in-NWs.

INTRODUCTION

Development of non-classical light generators utilizing the discrete density of states of quantum dots (QDs)[1] with high light-extraction efficiency has long been recognized as a major challenge in the context of quantum communication to enhance photon out-coupling to external optics. Many groups have demonstrated the enhancement of the light-collection efficiency from single photon sources in photonic nanowires (NWs) with carefully tailored end by applying a "top-down" approach using electron-beam (EB) lithography and reactive ion etching (RIE)[2,3]. Recently, formation of QD-in-NWs utilizing a "bottom-up" approach has been exploited as a promising candidate for such applications because of improved quality of surface morphology which is damaged by the top-down approach. Furthermore, the growth of QD-in-NWs on (111)-oriented substrates has attracted scientific attention for applications in entangled-photon emitters because fine-structure splittings induced by piezoelectricity, interface and strain asymmetries can be eliminated due to the threefold rotational symmetry of the (111) plane[4,5]. In addition, strain-free QDs would be obtainable in the growth of QD-in-NWs due to strain relaxation via lateral direction between lattice mismatched materials[6,7], which would enable the realization of ultra-high-efficiency intermediate-band solar-cells (IBSCs) utilizing densely-packed, multi-stack QDs[8]. In the formation of QD-in-NWs, there are predominantly two bottom-up approaches: the vapor-liquid-solid (VLS) method [9] and the "site-controlled", catalyst-free method using selective growth[10]. For high-performance NW-based optoelectronic devices, it is crucial to design and realize site-controlled QD-in-NWs in terms of size uniformity, position/orientation controllability, reproducibility and crystal quality due to their "catalyst-free" nature.

[1] E-mail: tatebaya@iis.u-tokyo.ac.jp

Many groups have so far reported the formation and optical properties of QD-in-NWs with various material systems including In(Ga)As/GaAs[11], GaAsP/GaP[12], AlGaAs/GaAs[13], InAsP/InP[14,15], and GaN/AlN[16]. For GaAs-based QD-in-NWs, several groups have investigated the formation of axial In(Ga)As/GaAs heterostructure NWs[11,17-19] and one of them has obtained the sharp excitonic peaks by using the VLS method[11]. However, there have been no reports of non-classical light generation from site-controlled In(Ga)As/GaAs QD-in-NWs. Dorenbos et al have reported the observation of photon anti-bunching and cascaded exciton-biexciton emission from site-controlled InAsP/InP QD-in-NWs[14]. It would be advantageous if site-controlled QD-in-NWs could be realized on a GaAs platform in terms of their compatibility with well-established, low-cost GaAs-based processing technologies for applications to SPEs and IBSCs. In this letter, we demonstrate the site-controlled formation of InAs/GaAs QD-in-NWs on patterned GaAs(111)B substrates by selective metalorganic chemical vapor deposition (MOCVD). Photon antibunching from InAs/GaAs QD-in-NWs are observed on a GaAs platform by means of auto-correlation measurement of the single InAs/GaAs QD, which provides evidence for single photon emission from high-quality QD-in-NWs.

EXPERIMENT AND DISCUSSION

All samples are grown by low-pressure MOCVD at a total pressure of 76 Torr on patterned semi-insulating GaAs(111)B substrates. Trimethylgallium (TMGa), trimethylindium (TMIn) and tertiarybutylarsine (TBAs) are used as Ga, In and As precursors, respectively. A 20-nm thick SiO2 growth mask is patterned with a 1-µm pitch and an average diameter of 40 nm using EB lithography and RIE. After the surface cleaning process with sulfuric acid for 2 minutes, wafers are loaded immediately into the reactor. All layers including GaAs core, InAs heterostructure and GaAs shell are grown at 750°C. First, GaAs core is grown by supplying TMGa and TBAs at a V/III ratio of 100. The precursor flow rates result in 0.5 Å/s equivalent planar growth rate on GaAs(100) substrates. Next, InAs heterostructure is formed by supplying TMIn and TBAs at a V/III ratio of 100 which is followed by the growth of a GaAs shell with a V/III ratio of 200. 5-sec growth interrupts are included before and after the formation of each layer to adjust the flux. Structural analyses of fabricated InAs/GaAs QD-in-NWs are carried out by using high-resolution scanning electron microscopy (SEM) and scanning transmission electron microscopy (STEM). Two different kinds of optical characterization are carried out at 10 K; one is a macro photoluminescence (PL) setup detected by an InGaAs nitrogen-cooled photodiode array under an excitation by diode-pumped solid-state lasers (532 nm) with a beam spot size of approximately 500 µm, and another is a confocal micro PL (µ-PL) setup under an excitation by continuous-wave (cw) Ti:Sapphire laser (813 nm) with a beam spot size of approximately 2 µm. The collected light from the single QD-in-NW is then sent to either a spectrometer equipped with a nitrogen-cooled Silicon charge-coupled device for single dot spectroscopy, or a Hanbury Brown and Twiss (HBT) type interferometer equipped with Si avalanche photodiodes for auto-correlation measurements.

Figure 1(a) shows the schematic and SEM images of fabricated GaAs NWs including InAs/GaAs QDs. High-uniform, site-controlled hexagonal GaAs NWs are formed with diameter and height of 69±2 nm and 1080±35 nm (aspect ratio of 15.8), respectively. Cross-sectional STEM images shown in Fig. 1(b) corroborate the existence of a single InAs/GaAs QD structure within the fabricated GaAs NW, as highlighted with dashed white lines. Black-and-white contrast in the vicinity of the QD is indicative of the existence of strain fields in the matrix.

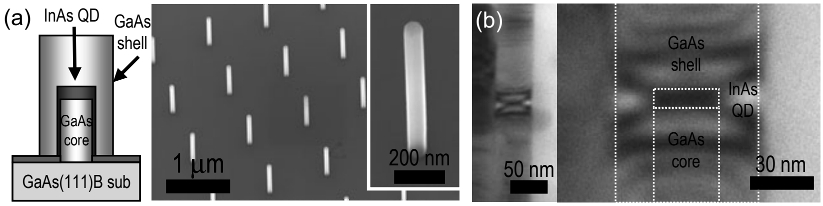

Fig. 1 (a) Schematic and SEM images of InAs/GaAs QD-in-NWs grown on patterned GaAs(111)B substrates. (b) Cross-sectional STEM images of a single InAs/GaAs QD-in-NW with diameter and height of 35 and 4.5 nm, respectively.

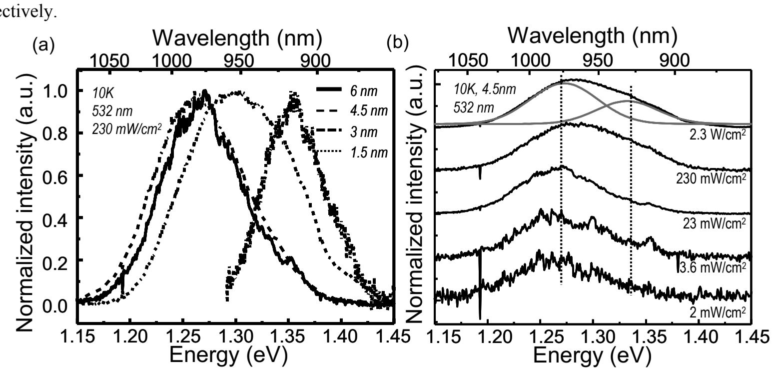

Fig. 2 (a) Macro PL spectra at 10K of InAs QD-in-NWs with InAs heights of 1.5, 3, 4.5, and 6 nm, and peak emission energies of 1.36, 1.30, 1.26, and 1.27 eV, respectively. (b) Macro PL spectra at 10 K of InAs/GaAs QD-in-NWs with a QD height of 4.5 nm at various incident pump-power densities from 2 mW/cm^2 to 2.3 W/cm^2.

Thickness and diameter of the observed InAs/GaAs QD are approximated to be 4.5 and 35 nm, respectively. Figure 2(a) shows the dependence of the normalized PL properties of InAs/GaAs QD-in-NWs at various thicknesses, which exhibits a blueshift of PL peaks from 1.27 to 1.36 eV with decreased thicknesses of InAs/GaAs heterostructures from 6 to 1.5 nm, which is attributed to the enhancement of quantum confinement effect along the vertical direction. It is noted that the PL peak energies of InAs/GaAs QD-in-NWs are much larger than those of conventional self-assembled InAs/GaAs QDs emitting in the fiber-optic communication regime (<1 eV)[20]. This is due to interdiffusion of In and Ga adatoms during the growth of QD-in-NWs at a comparatively high temperature. Figure 2(b) shows the typical PL spectra of InAs/GaAs QD-in-NWs with a height of 4.5 nm at various pump-power densities from 2 mW/cm^2 to 2.3 W/cm^2. Only a single peak is observed at 1.27 eV with a full-width at half-maximum (FWHM) of 92 meV at a very low pump-power density. The large FWHM is attributed to size distribution and structural fluctuation of InAs/GaAs QD-in-NWs and further optimization of the growth conditions of the QD formation as well as pattern processing is required to obtain more uniform QD-in-NWs. By contrast, at pump-power densities higher than 230 mW/cm^2, another peak emerges at 1.33 eV which likely originates from higher subband of the QDs. These results indicate that the observed PL peaks originate from fabricated InAs/GaAs heterostructures.

Single dot spectroscopy of a single InAs/GaAs QD-in-NW is carried out by using μ-PL characterization setup. The emission spectrum displays several sharp lines with cw excitation

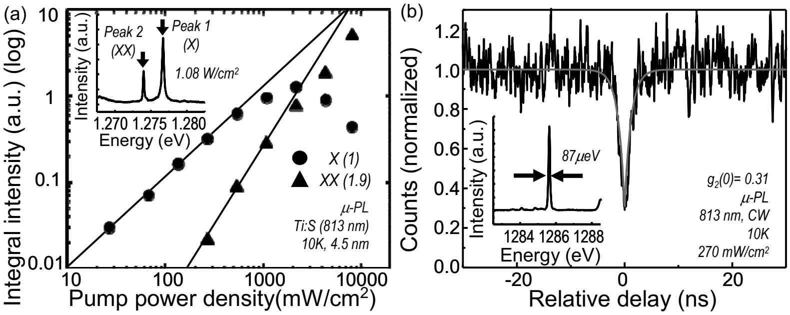

Fig. 3 (a) A log-log plot of integrated PL intensities of a single InAs/GaAs QD-in-NW at 10 K versus pump-power densities from 14 mW/cm^2 to 16.8 W/cm^2. An inset shows typical μ-PL spectrum at 10 K of a single InAs/GaAs QD-in-NW at a pump-power density of 1.08 W/cm^2. (b) Auto-correlation histogram of an exciton energy peak from a single InAs/GaAs QD-in-NW with a linewidth of 87 μeV under cw excitation as a function of the relative delay. A solid gray line shows a fitting curve. An inset shows the μ-PL spectrum of the single InAs/GaAs QD-in-NW used for the auto-correlation measurement.

above the GaAs band gap. Figure 3(a) shows a log-log plot of integrated PL intensity versus pump power densities from 14 mW/cm^2 to 16.8 W/cm^2 of two dominant peaks from a single QD-in-NW as shown in the inset. At very low pump power densities, only a single peak (peak 1) is observed at the peak energy of 1276.7 meV with a linewidth of 393 μeV and exhibits linear dependence against pump power densities. By increasing pump power densities, another peak (peak 2) emerges at the peak energy of 1274.0 meV which shows quadratic dependence. These results indicate that observed sharp peaks 1 and 2 originate from exciton and biexciton emissions, respectively. The obtained linewidth of a single exciton peak varies from <100 to several hundred μeV which is attributed to the size distribution of InAs QDs in GaAs NWs. These values are almost comparable to those of QD-in-NWs reported by other groups, but larger than those of conventional SK QDs, which is due to spectral diffusion caused mainly by surface states of GaAs NWs that traps and detraps carriers. Further investigation and analyses are required to corroborate the effect of surface states on the μ-PL characteristics of a single InAs QD-in-NW.

Measurement of the second-order coherence function, $g^{(2)}(\tau)$, of the QD-in-NWs with the narrowest linewidth of 87 μeV as shown in the inset of Fig 3(b) is carried out at 10 K to corroborate the existence of the fully-quantized energy levels in the single QD-in-NW structure. Figure 3(b) show auto-correlation histogram of the normalized coincidence counts of exciton emission from a single QD-in-NW at the pump power density of 420 mW/cm^2. The obtained auto-correlation, $g^{(2)}(\tau)$, in Fig. 3(b) is fitted by $g^{(2)}(\tau)= 1-(1-g^{(2)}(0))\exp(-|\tau|/\tau_0)$ to extract the fitting parameters, $g^{(2)}(0)$ and τ_0. The second-order coherence function shows a large degree of photon antibunching with a fitting parameter $g^{(2)}(0)= 0.31$, which clearly proves the existence of fully-quantized energy states in the InAs/GaAs QD-in-NW.

CONCLUSIONS

We report the demonstration of site-controlled InAs/GaAs QD-in-NWs on patterned GaAs(111)B substrates by selective MOCVD. PL peaks from InAs QD-in-NWs are identified by varying the thickness of InAs heterostructures using a macro-PL characterization at 10 K. STEM analyses corroborate the existence of a single InAs/GaAs QD structure embedded in a GaAs NW and approximate the dimension of the InAs/GaAs QD-in-NW. Sharp excitonic peaks from a single InAs/GaAs QD-in-NW are observed using a μ-PL setup at 10K. The exciton linewidth is as narrow as 87 μeV. Auto-correlation measurements of a single InAs/GaAs QD-in-NW using HBT-type configuration shows a photon antibunching with $g^{(2)}(0)$ of 0.31. These results provide evidence for single photon emission from fully-quantized energy levels in high-quality QD-in-NWs, which would enable the implementation of high-performance optoelectronic devices based on site-controlled QDs, including SPEs and IBSCs, utilizing existing, well-established NW growth technologies.

ACKNOWLEDGMENTS

This work is supported by the Special Coordination Funds for Promoting Science and Technology and Funding Program for World-Leading Innovative R&D on Science Technology. The authors would like to acknowledge Dr. T. Ishida, Ms. Y. Takayama and Prof. M. Fujita for TEM analyses of fabricated InAs/GaAs QD-in-NWs.

REFERENCES

1. Y. Arakawa and H. Sakaki, Appl. Phys. Lett. 40, 939 (1982).
2 T. M. Babinec, B. J. M. Hausmann, M. Khan, Y. Zhang, J. R. Maze, P. R. Hemmer and M. Lončar, Nature Nanotechnology 5 (2010) 195.
3 J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne and J. -M. Gérard, Nature Photon. 4 (2010) 174.
4 R. Singh and G. Bester, Phys. Rev. Lett. 103, (2009) 063601.
5 A. Schliwa, M. Winkelnkemper, A. Lochmann, E. Stock, and D. Bimberg, Phys. Rev. B 80, (2009) 161307.
6 E. Ertekin, P. A. Greaney, D. C. Chrzan, and T. D. Sands, J. Appl. Phys. 97, 114325 (2005).
7 F. Glas, Phys. Rev. B, 74, 121302 (2006).
8 T. Nozawa and Y. Arakawa, Appl. Phys. Lett. 98, 171108 (2011).
9 R. S. Wagner, and W. C. Ellis, Appl. Phys. Lett. 4, 89 (1964).
10 J. Motohisa, J. Noborisaka, J. Takeda, M. Inari, and T. Fukui, J. Cryst. Growth 272, 180 (2004).
11 N. Panev, A. I. Persson, N. Sköld, and L. Samuelson, Appl. Phys. Lett. 83, 2238 (2003).
12 M. T. Borgström, V. Zwiller, E. Müller, and A. Imamoglu, Nano Lett. 5, 1439 (2005).
13 H. Sanada, H. Gotoh, K. Tateno and H. Nakano, Jpn. J. of Appl. Phys. 46, 2578 (2007).
14 S. N. Dorenbos, H. Sasakura, M. P. van Kouwen, N. Akopian, S. Adachi, N. Namekata, M. Jo, J. Motohisa, Y. Kobayashi, K. Tomioka, T. Fukui, S. Inoue, H. Kumano, C. M. Natarajan, R. H. Hadfield, T. Zijlstra, T. M. Klapwijk, V. Zwiller, and I. Suemune, Appl. Phys. Lett. 97, 171106 (2010).
15 M. E. Reimer, G. Bulgarini, N. Akopian, M. Hocevar, M. B. Bavinck, M. A. Verheijen, E. P. A. M. Bakkers, L. P. Kouwenhoven and V. Zwiller, Nature Comms, 3:739 (2012).
16 J. Renard, R. Songmuang, C. Bougerol, B. Daudin and B. Gayral, Nano Lett. 8, 2092 (2008).
17 M. Paladugu, J. Zou, Y.-N. Guo, X. Zhang, Y. Kim, H. J. Joyce, Q. Gao, H. H. Tan and C. Jagadish, Appl. Phys. Lett. 93, 101911 (2008).
18 M. Heiß, A. Gustafsson, S.Conesa-Boj, F. Peiró, J. R. Mo-rante, G. Abstreiter, J. Arbiol, L. Samuelson and A. F. Morral, Nanotechnology 20, 075603 (2009).
19 J. N. Shapiro, A. Lin, P. S. Wong, A. C. Scofield, C. Tu, P. N. Senanayake, G. Mariani, B. L. Liang and D. L. Huffaker, Appl. Phys. Lett. 97, 243102 (2010).
20 J. Tatebayashi, M. Nishioka, and Y. Arakawa, Appl. Phys. Lett. 78, 3469 (2001).