Nano World Journal

https://doi.org/10.17756/nwj.2023-s5-040

Research Article

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Polarization Dependence of Excitonic Emission from As-rich Single $InAs_xP_{1-x}$ Quantum Dot Embedded in Free-standing InP Nanowire

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Received: October 11, 2023
Accepted: December 20, 2023
Published: December 22, 2023

Citation: Mukherjee S, Tomioka K, Motohisa J. 2023. Polarization Dependence of Excitonic Emission from As-rich Single $InAs_xP_{1-x}$ Quantum Dot Embedded in Free-standing InP Nanowire. *NanoWorld J* 9(S5): S202-S205 .

Published by United Scientific Group

Abstract

Position-controlled MOVPE (metal-organic vapor phase epitaxy) growth of free-standing InP nanowire (NW) with embedded $InAs_xP_{1-x}$ quantum dots (QD) having a large As composition is reported here. Impression of exciton and bi-exciton emission from individual $InAs_xP_{1-x}$ QD has been confirmed by micro-photoluminescence (μ-PL) measurements of single standing NW-QD and by studying the excitation intensity dependence of the PL-spectra. Fine structure spitting of exciton emission confined in As-rich QD has been quantitatively determined from polarization dependent μ-PL measurements of single standing NW-QD heterostructure.

Keywords

Selective area growth, Exciton fine structure, Quantum dot, Nanowire

Introduction

Recently, semiconductor NW-QD integrated heterostructure with QD embedded inside the NW has opened up new possibility to integrate QDs to modern semiconductor electronics [1]. This integrated system is a promising candidate for future low loss single and entangled photon sources for quantum information applications [2]. The inherent alignment of QDs along the NW long axis offers a natural 1D channel for the charge carriers and high light extraction efficiency with easily making electrical contact on single vertical NW [3]. This brings in flexibility on designing novel optoelectronic devices and transmitting quantum information systems [4].

Individual semiconductor QD embedded in NW is a popular choice for producing polarization-entangled two-photon source, provided the spins of the first photon steaming from bi-exciton (|XX> to the intermediate degenerate exciton (|X>) state and the second photon coming from the X-state to the ground state (|0>) are entangled [5]. This entanglement requires a vanishing excitonic fine structure splitting (FSS) which normally breaks down in presence of finite e-h anisotropic exchange interaction that leads to non-zero X-FSS [6]. The in-plane geometry of NW-QD system plays a vital role degerming the interaction. The X-FSS is mainly attributed to lateral shape anisotropy of the QD-NW system or some piezoelectric field due to strain [7]. Exciton FSS also depends on the orientation of the grown NW over the substrate. Some group theoretical calculations predict the FSS can be minimized if the growth takes place along [111] direction over (111)-oriented substrate [5].

It has been quite a while, the bright exciton emission confined in QDs of $InAs_xP_{1-x}$ embedded in InP NW has been studied as a source of single photons for 1D laser and other quantum cryptographic applications [8]. But to have QD excitonic light emission in telecom wavelength range (1.25 - 1.6 μm), the embedded $InAs_xP_{1-x}$ layer should be rich in As content (x). But the redshift of PL line only comes at the cost of size and shape anisotropy of the grown QDs and incoherent relaxation of large lattice strain in the $InAs_xP_{1-x}$/InP interface [9]. These are the possible attributes at large As composition that can lead to large exciton fine structure splitting and degrade its potential as source of entangled-photon pairs. This limits the study of QD bright-excitonic single photon emission in $InAs_xP_{1-x}$/InP QD-NW heterostructure to lower As concentration to minimize X-FSS. But it is worth studying the bright exciton emission from single As-rich $InAs_xP_{1-x}$/InP QD-NW at longer wavelength (1.25 - 1.6 μm). It can indeed come handy to look for improvised design and optimized growth engineering with the plausibility of lowering the X-FSS for future telecommunication applications. This has motivated us to study the bright X-emission from single rather less explored As-rich $InAs_xP_{1-x}$ QD embedded in single standing InP NW under optimized growth condition and have quantitatively determined the X-FSS.

Here we report selective-area MOVPE growth of vertical InP/$InAs_xP_{1-x}$/InP NW heterostructure containing embedded $InAs_xP_{1-x}$ QDs with large As composition. Impression of exciton and bi-exciton emission from individual InAsP QD has been confirmed by μ-PL measurements of single standing NW-QD and by studying the excitation intensity dependence of the PL-spectra. Fine structure spitting of exciton emission has been quantitatively determined from polarization dependent μ-PL measurements of single standing NW-QD.

Experimentation

Sample preparation: array of standing NW-QD heterostructure

Array of free-standing InP/$InAs_xP_{1-x}$/InP vertical NW heterostructures with embedded $InAs_xP_{1-x}$ QDs have been grown on patterned InP (111)-A substrate by selective area MOVPE, as shown in figure 1. At first, InP (111)-A substrate has been partially covered with SiO_2 of thickness 30 nm by radio frequency sputtering. Thereafter, the nucleation sites for the growth of InP NWs are generated in the form of a periodic array of 80 - 100 nm diameter masked pattern of pitch 3 μm, created by electron beam lithography followed by wet-etching. Then, a long vertical segment of InP has been grown over the openings on SiO_2 by chemically reacting Gr-III and V precursors trimethylindium (TMI) and tertiary butyl phosphine (TBP) for 15 min in the MOVPE reactor maintained at 660 °C. Subsequently, the reactor temperature has been lowered to 580 °C for QD growth. A thin layer of $InAs_xP_{1-x}$ has been grown by adding arsine (AsH_3) along with TBP and TMI in the reactor for 3 s with a V/III ratio of 520 at a rich As background with Gr-V partial pressure of AsH_3/TBP = 1.5, where x is the As composition. Subsequently, the grown QDs are capped by a layer of InP before the reactor temperature again increased from 580 °C to 660 °C. Finally, the QDs are embedded under a second long vertical segment of InP grown at 660 °C.

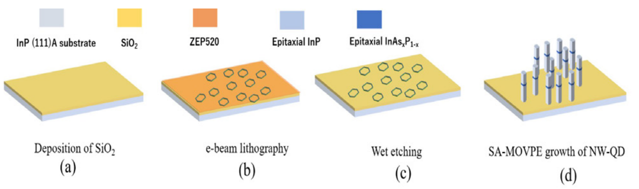

Figure 1: Schematic representation of selective area MOVPE growth of free-standing InP/InAsP/InP NW-QD heterostructure on patterned (111)-A InP substrate.

Vertically oriented periodic array of 1.3 μm long (average) InP/$InAs_xP_{1-x}$/InP NW heterostructure of average diameter of 110 nm have been grown with a pitch (period) length of 3 μm, as shown in the SEM (scanning electron microscope) image (Figure 2).

Experimental set up: optical spectroscopy

A standard μ-PL measurement of the as-grown InP/$InAs_xP_{1-x}$/InP NW heterostructure attached to (111)-A InP substrate have been carried out over the temperature range of 4.2 K to 300 K. Individual NWs mounted inside a He-flow cryostat have been excited along NW long c-axis by continuous 632.8 nm He-Ne laser focussed to 2 μm spot size over the sample surface by using a 0.42 N.A. microscope objective (50X). PL from individual NWs have been collected by the same microscope objective and fed into a half-waveplate, followed by a fixed infrared linear polarizer for determination of the state of linear polarization of the emitted light. The half-waveplate has been rotated to access different polarization state of the PL collected from single NW. Then the linear polarized PL signal from the polarizer has been collected and dispersed with a double (f = 1.0 m) grating spectrometer and

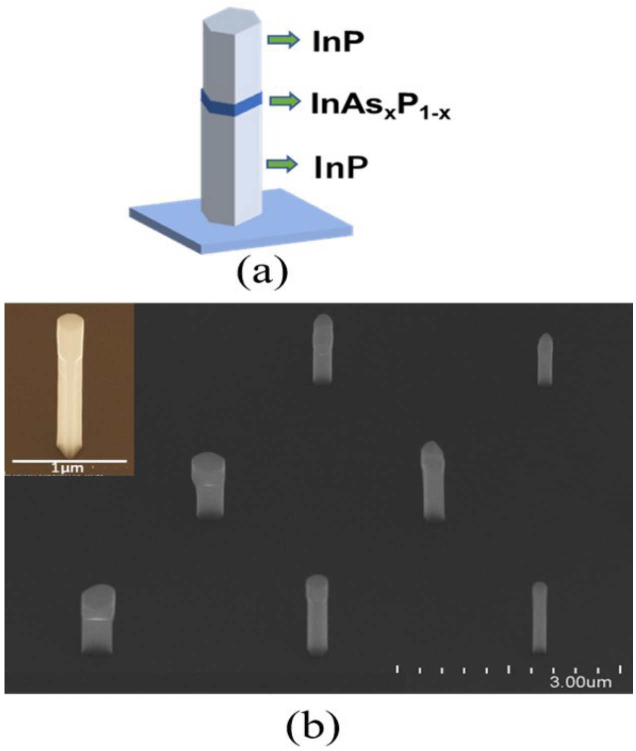

Figure 2: **(a)** Schematic representation and **(b)** SEM image of the standing InP NW with embedded InAsP QD grown on (111)-A InP patterned substrate.

subsequently detected by a liquid nitrogen cooled InGaAs photodiode array detector.

Results and Discussion

The μ-PL spectrum of single standing $InP/InAs_xP_{1-x}/InP$ NW-QD heterostructure excited by the saturation excitation intensity is shown in figure 3. Figure 3a shows PL spectrum of QD excited with excitation intensity which saturates the ground state transitions in $InAs_xP_{1-x}/InP$ QW, identified as emission line from neutral exciton (X) and bi-exciton (XX) recombination at around 0.837 eV and 0.842 eV from excitation power dependent PL measurements (not shown here). The binding energy of bi-exciton $\left(E^X - E^{XX} = -5\text{meV}\right)$ has come out to be negative which is most likely to be attributed to strong Coulombic interaction between the holes in XX bound state, more common in small sized QDs [10].

The PL peak around 1.49 eV (Figure 3b) corresponding to band-to-band recombination from WZ InP [11]. The energy corresponding to strained $InAs_xP_{1-x}/InP$ QW radiative transition (c-hh) from excited e-state in CB to hh-state in VB due to close-to-bandgap c-hh transition can be written as [11-13].

$$E_{c-hh}(x) = E_g^{InAs_xP_{1-x}} + 3a_{ph}\left(2\in_{XX} + \in_{ZZ}\right) + E^{conf} \quad (1)$$

The compressive in-plane strain acting on the embedded $InAs_xP_{1-x}$ layer plays a very significant role on the optical properties and leads to a PL peak red shift. The strain effect has been accommodated by the 2nd term in (1), where a_{ph} is the phonon deformation potential [14, 15], ϵ_{zz} is the out-of plane strain,

$$\varepsilon_{zz} = -2\left(C_{13}/C_{33}\right)\varepsilon_{xx} \quad (2)$$

under the approximation of uniaxially strained [111]-oriented pseudomorphic growth of $InAs_xP_{1-x}$ over lattice-mismatched WZ (hexagonal lattice) InP with isotropic in-plane strain

$$\varepsilon_{xx} = \varepsilon_{yy} = \left(a^{InAs_xP_{1-x}} - a^{InP}\right)/a^{InAs_xP_{1-x}} \quad (3)$$

over the rotationally symmetric xy plane [16]. The lattice constant of $InAs_xP_{1-x}$ has been estimated as a function of x, by linearly extrapolating the lattice constants of bulk InAs and InP [14]. The sum over the confinement energy E^{conf} E^{conf} of e-h pair trapped in QD's confining potential, has been accommodated in the 3rd term of (1).

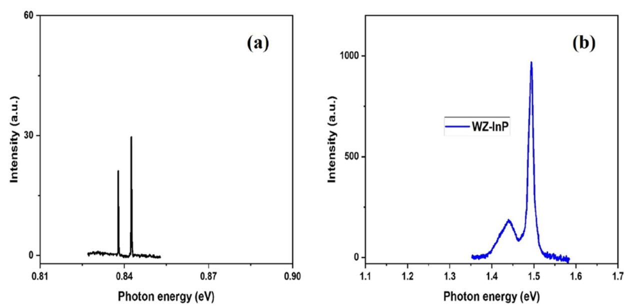

Figure 3: **(a)** μ-PL spectrum of single standing $InP/InAs_xP_{1-x}/InP$ NW-QD heterostructure excited by the saturation excitation intensity and **(b)** The PL peak at 1.49 eV due to band-to-band transition from WZ InP.

Polarization dependent PL measurement: exciton fine structure splitting

The shift in exciton (X) peak position for varying direction of polarization of the collected PL signal has been obtained by rotating a half-waveplate just at the back of a linear polarizer in the entrance of the spectrometer. The direction of polarization of the collected PL signal has been specified by polarization angle (θ) measured with respect to some fixed direction (θ_0). The measured energy of the X peak is plotted for different values of θ, as shown in figure 4. The polarization dependence of X-peak energy has been modelled by standard sine function $E^X(\theta) = E_0 + A\sin\left[2(\theta - \theta_0)\right]$, where is the offset angle and A and are positive fitting constants. The peak-to-peak energy difference this fitting curve is taken to be the measure of the fine structure splitting of excitonic transition which has been determined to be 76 ± 2 μeV [11].

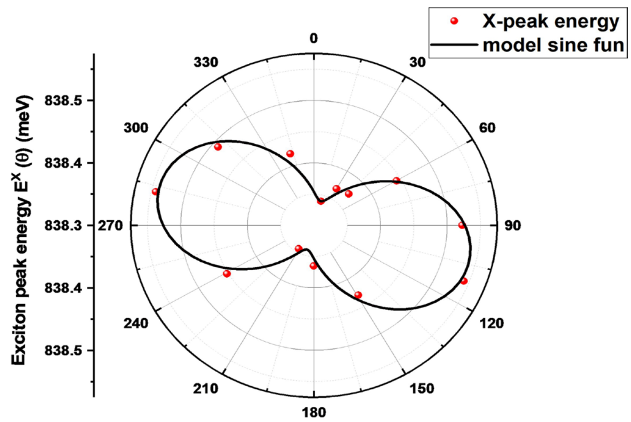

Figure 4: The variation of measured energy (shown by red dot) of the X-PL peak is plotted for different values of angle of polarization θ and modelled by standard sine-function with offset value (modelling shown by solid line).

Conclusion

Array of vertically oriented $InP/InAs_xP_{1-x}/InP$ QD-NW heterostructure have been grown on patterned (111)-A InP substrate with single QD in each NW by selective area-MOVPE. The existence of exciton and bi-exciton emission originated from the embedded QD has been confirmed from μ-PL measurements of single QD-NW heterostructure and the excitation intensity dependence of the PL spectrum. We have measured a nonvanishing moderate exciton FSS for InAsP QD embedded in the [111]-oriented InP NW by studying the polarization dependence of the PL spectra of single QD-NW. This is attributed to a (substantial) anisotropic exchange interaction, most probably attributed to non-zero piezoelectric field induced by large lattice mismatch or lateral QD elongation. Finally, with sharp QD exciton emission line, this single QD-NW heterostructure excites us to measure its single photon emission probability which is under way and planned to be reported afterwards.

Acknowledgements

The authors would like to thank Prof. Agus Subagyo for fruitful discussion with him.

Conflict of Interest

None.

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