

Exciton and biexciton emissions from single GaAs quantum dots in (Al,Ga)As nanowires

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1. Introduction

Exploring more accurate ways of controlling the shapes and positions of semiconductor nanostructures is a major challenge as regards the realization of such future applications as quantum information devices. In the last decade, it has been reported that heterojunctions can be introduced in semiconductor nanowires (NWs) via vapor-liquid-solid (VLS) crystal growth [1-6]. Furthermore, a few groups have recently observed exciton emissions from single quantum dots (QDs) in VLS-grown NWs fabricated using $In_xGa_{1-x}As$ [7] or $GaAs_{1-x}P_x$ [8] compounds. In addition to these strained heterostructures, we expect a lattice-matched $Al_xGa_{1-x}As$ system to be important in terms of both flexible material choice and the length-scale design of the nanostructures. In this report, we present the results of an experimental photoluminescence (PL) measurement for GaAs QDs embedded in (Al,Ga)As NWs grown by the VLS method. These results show that our method is a promising technique for obtaining controllable nanostructures.

2. Experimental

The sample was grown on a GaAs (311)B substrate in a low-pressure metal-organic vapor phase epitaxy (MOVPE) reactor. To embed the QDs in the NWs, we combined VLS growth and normal MOVPE growth as follows [9]. First, we formed small Au nano-particles as catalysts (diameter ~ 25 nm, density $d = 30\text{-}50 \mu\text{m}^{-2}$) by annealing Au film deposited on the substrate. The $Al_xGa_{1-x}As$ NWs were grown along the [111]B direction in the VLS growth mode in a trimethylgallium (TMG), trimethylaluminum (TMA) and AsH_3 atmosphere [9]. The substrate temperature was set at $T_s = 470^\circ C$. We programmed the TMG and TMA supplies sequentially so that cylindrical GaAs QDs approximately 20 nm in length were formed between the $Al_{0.3}Ga_{0.7}As$ NWs [10]. Finally, the surfaces of the NWs were covered with 25 nm-thick $Al_{0.4}Ga_{0.6}As$, 25 nm-thick $Al_{0.6}Ga_{0.4}As$, and 5 nm-thick GaAs films by using normal MOVPE growth at $T_s = 600\text{-}700^\circ C$. After the growth, we randomly scattered the NWs on a Si(100) substrate ($d \sim 0.1 \mu\text{m}^{-2}$) in order to measure spatially-separated single NWs.

To investigate the optical quality of our sample, we carried out a micro-PL measurement at low temperature. The sample was cooled to $T = 4.14$ K in a liquid helium-flow cryostat with an optical window. We used a cw Ti:sapphire laser for the excitation and it was focused on a single NW

with a spot size of about 2 μm by using an objective lens. The PL from single NWs was dispersed through a spectrometer and detected by a charge coupled device camera. Linear polarizers and waveplates were installed in this micro-PL system, so that the linear polarization axis of the excitation laser and the detected component of the PL polarization were in the same direction. We adopted the relative angle between this optical polarization axis and the sample (i.e. Si substrate) axis as a parameter, θ .

3. Results and discussion

The PL spectra observed with different excitation powers (P_{exc}) are plotted in Fig. 1(a). At a moderate P_{exc} , we found two peaks at $E = 1.671$ eV (Peak A) and 1.668 eV (Peak B) with line widths of 1-2 and 3-10 meV, respectively. Since both peaks are well fitted by single Lorentz functions, these signals may be emitted from a single QD. The fact that the peak energies are much higher than the GaAs bandgap indicates the possibility that the QDs are composed of $Al_xGa_{1-x}As$ rather than pure GaAs. As P_{exc} increases, Peak A saturates at $P_{\text{exc}} \sim 100 \text{ mW/cm}^2$, while Peak B increases with the red shift and becomes larger than Peak A. In Fig. 1(b), we plot the integrated PL intensities of Peaks A and B as a function of P_{exc} . The slopes of Peaks A and B are well fitted by $N = 1$ and 2. This behavior is characteristic of exciton and biexciton emissions. It is unclear why the line width of Peak B is much larger than that of

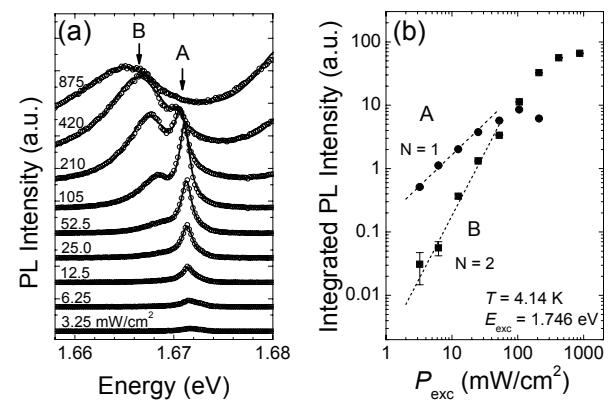


Fig. 1 (a) PL spectra of a single QD in a NW for several excitation powers at $T = 4.14$ K. The lines are fitted to the data with a Lorentz function. (b) Integrated PL intensities of the two main peaks as a function of excitation intensity.

Peak A and the energy shifts to a lower value, but a similar feature has also been observed for $\text{In}_x\text{Ga}_{1-x}\text{As}$ [7] and $\text{GaAs}_{1-x}\text{P}_x$ [8] QDs in NWs.

In Fig. 2, we plot the polarization dependence of the PL intensities to evaluate the three-dimensional shapes of the QDs. The intensity of Peak A has its maximum value at $\theta = 100^\circ$ and decreases sharply at $\theta = 10^\circ$. Note that the angle for the maximum intensity is not always the same, because the NWs are randomly glued to the Si substrate. We define the polarization P as,

$$P (\%) = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \times 100, \quad (1)$$

where I_{\max} (I_{\min}) is the maximum (minimum) PL intensity. The experimental value of $P = 95\%$ results from the selectivity of the linear polarization in the excitation (50 %) and emission process (80 %). Such a strong dependence of the PL intensity on optical polarization might be due to the highly asymmetrical shape of the present QDs; probably ‘disk-like QDs’ or ‘wire-like QDs’ in our case. The result ensures that a nanostructure with large optical anisotropy is formed by the VLS method.

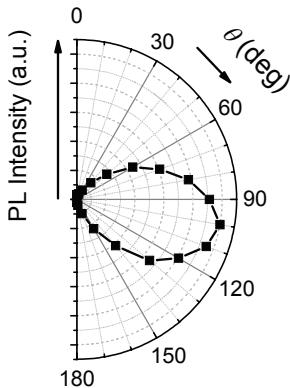


Fig. 2 Optical polarization angle dependence of the PL intensity at $E = 1.671$ eV. θ is the relative angle between the optical polarization axis and the sample.

4. Summary

We have shown that it is possible to produce optically active QDs in VLS-grown NWs using the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ system. The PL from a single QD in a NW exhibits exciton and biexciton emission behavior. The intensity of the exciton emission depends on the optical polarization axis, indicating the highly asymmetrical shapes of the QDs. The VLS method is a good candidate with which to obtain nanostructures whose optical and electronic properties can be designed.

Acknowledgements

We thank H. Kamada, T. Sogawa, H. Yamaguchi, Y. Tokura, and M. Morita for their continuous encouragement throughout this work. This work was partly supported by Japan Society for the Promotion of Science (JSPS).

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