Research Paper

Photoluminescence Characterization of Vertically Coupled Low Density InGaAs Quantum Dots for the application to Quantum Information Processing Devices

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Abstract Vertically coupled low density InGaAs quantum dots (QDs) buried in GaAs matrix were grown with migration enhanced molecular beam epitaxy method as a candidate for quantum information processing devices. We performed excitation power-dependent photoluminescence measurements at cryogenic temperature to analyze the effects of vertical coupling according to the variation in thickness of spacer layer. The more intense coupling effects were observed with the thinner spacer layer, which modified emission properties of QDs significantly. The low surface density of QDs was observed by atomic force microscopy, and scanning transmission electron microscopy verified the successful vertical coupling between low density QDs.

Keywords: Quantum dots, Molecular beam epitaxy, InGaAs, photoluminescence, Quantum information processing

I. Introduction

The low dimensional semiconductor nanostructures are promising material systems for novel photonic devices. Especially, the zero-dimensional structures such as quantum dots (QDs) have been of great interest for quantum information processing devices, *e.g.* single photon sources [1,2] and quantum gates [3]. Among many other possible materials, In(Ga)As QDs are favorable for the realization of quantum information processing devices due to matured growth technology. As the emission wavelength range of In(Ga)As QDs falls in near-infrared (NIR) region, including the wavelengths employed in modern optical fiber communication systems, devices made of such QDs can be readily integrated [4]. Especially, vertically coupled pairs of In(Ga)As QDs are promising for quantum information processing [5].

To utilize such quantum mechanically coupled QDs, it is essential to characterize the effect of the coupling. For this reason, we have grown samples of two InGaAs/GaAs QD layers with different coupling distances by migration enhanced molecular beam epitaxy (MEMBE) method which permits uniform size and low surface density. The low density of QDs is beneficial for the optical characterization of a single QD or a single pair of vertically coupled QDs because the lower density allows for less

degree of interference between nearby QDs [6,7]. We analyzed the optical properties of InGaAs QDs, especially focused on the effects of vertical coupling, by performing excitation power-dependent photoluminescence (PL) measurements at cryogenic temperature.

II. Experiments

The low density InGaAs QDs buried in GaAs barriers were grown by MEMBE method [8,9], with long migration enhancing time to obtain low surface density of QDs [10].

GaAs 5 nm
Al _{0.3} Ga _{0.7} As 20 nm
GaAs 20 nm
InGaAs low density QDs
GaAs spacer layer (5 nm, 8 nm, 10 nm)
InGaAs low density QDs
GaAs 20 nm
Al _{0.3} Ga _{0.7} As 1 nm/ GaAs 1 nm SPS (X5)
Al _{0.3} Ga _{0.7} As 50 nm
GaAs buffer
S.I. GaAs

Figure 1. The structure of vertically coupled low density InGaAs/GaAs QD samples grown by MEMBE method.

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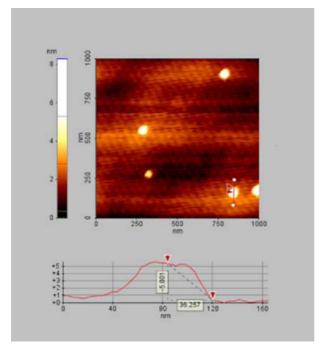


Figure 2. The AFM image analysis of low density InGaAs QDs.

To enhance the surface flatness of a substrate, the QD layers were grown on a structure consists of five pairs of short-period-superlattice (SPS) (each pair consists of a 1 nm-thick Al_{0.3}Ga_{0.7}As layer and a 1 nm-thick GaAs layer). We fabricated three samples with two layers of InGaAs QDs separated by a GaAs spacer layer (SL), where the thickness of SL was varied as 10 nm, 8 nm, and 5 nm (Fig. 1). For comparison, a sample with only single layer of QDs was grown as well. These samples are denoted as SL 10 nm, SL 8 nm, SL 5 nm, and SG, respectively.

We characterized the morphology and surface density of InGaAs QDs using an atomic force microscopy (AFM) in contact mode with a contact-type cantilever which has tip curvature radius of <10 nm (Fig. 2). Additionally grown samples without capping layers on top of QDs were used

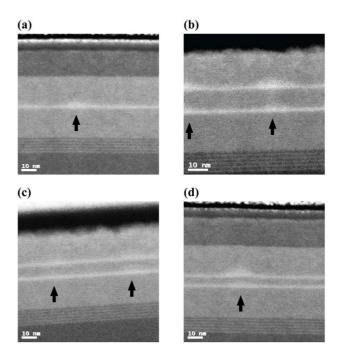


Figure 3. The STEM image analysis of coupled InGaAs QD layers: (a) SG, (b) SL 10 nm, (c) SL 8 nm, and (d) SL 5 nm.

for this purpose to permit the AFM tips to be accessible to the QDs. The surface density of QDs was estimated from averaging the number of QDs within $1 \mu m^2$ area of each image, and the average dimensions of QDs were identified through analyzing the line profiles of each QD. The measured AFM images revealed that the InGaAs QDs grown by MEMBE method exhibit high uniformity in size and low surface density of $\sim 5 \mu m^{-2}$. The average height and width of QDs were $\sim 4 \mu m$ and $\sim 70 \mu m$, respectively.

To obtain the precise coupling distances between two InGaAs QD layers, we measured cross-sectional scanning transmission electron microscopy (STEM) images (Fig. 3), where cross-section specimens were prepared by focused ion beam milling. The brighter InGaAs QD layers are clearly distinguishable from the surrounding darker GaAs

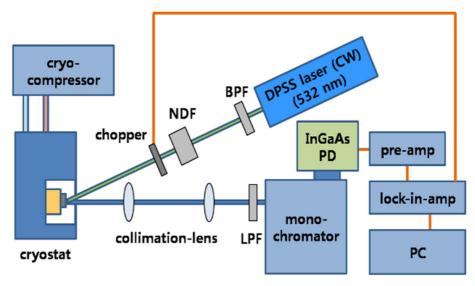


Figure 4. The cryogenic temperature PL measurement setup.

layers, and the position of QDs are clarified by the black arrow markers below them in the STEM images (Also, the Al_{0.3}Ga_{0.7}As/GaAs SPS layer is clearly visible at the bottom of each image.) Through the analysis of these images, the actual thickness of SL for each sample was determined: for samples of SL 10 nm, SL 8 nm, and SL 5 nm, the measured thickness values were 12.0 nm, 9.3 nm, and 6.7 nm, respectively. For all range of different SL thickness values, the successful vertical alignment of two self-assembled InGaAs QD layers were observed. Due to low density of QDs, the observation of QDs during the TEM measurement is dependent on the probability. However, the vertical coupling between two layers of QDs is well observed if the QDs are found. It is attributed to the relatively thin thickness of SL to transfer the strain from the lower layer to the upper layer of the QDs and long migration time to let the upper QDs find the strain from the lower QDs during QD formation.

The PL measurement setup is illustrated in Fig. 4. The diode-pumped solid state (DPSS) continuous wave (CW) laser (λ =532 nm) was the excitation source. A band pass filter (BPF) was used to remove out-of-band wavelength components from the laser output, and a set of neutral density filters (NDF) was utilized to adjust the power of laser beam. At the entrance slit of monochromator, a long pass filter (LPF) blocks scattered laser light from the sample surface. The emission from a sample was collimated and dispersed in the monochromator with a grating of 600 mm⁻¹ ruling and 1000 nm blaze wavelength. The dispersed light was detected from Peltier-cooled InGaAs photodiode (PD) and then amplified through preamplifier and lock-in amplifier combined with optical chopper to enhance signal to noise ratio. The measurement temperature was kept at 15 K with closed cycle Helium cryostat.

III. Results and Discussion

Firstly, samples were measured at 15 K with fixed excitation power of 70 mW and shown as normalized PL spectra for comparison (Fig. 5). The samples of two vertically coupled QD layers (SL 10 nm, SL 8 nm, and SL 5 nm) exhibit distinct multi-peak shapes, while the sample of a single QD layer (SG) has a single dominant peak. For a smaller SL thickness, the multi-peak shape becomes more obvious, which indicates stronger degree of vertical coupling between QDs [11,12]. The degree of coupling is nearly proportional to the amount of red-shift in the highest peak position of each vertically coupled QD sample, compared to the peak position obtained from a single QD layer (SG). Actually, decreasing SL thickness (from 10 nm to 8 nm, and then to 5 nm) induced more significant amount of red-shift (93.0 meV, 103.3 meV and 123.3 meV, respectively). This effect can be attributed to the reduction of ground state energy due to mini-band formation caused by

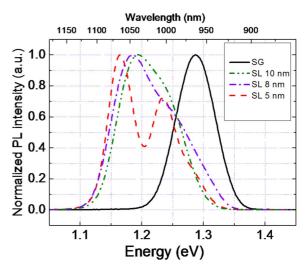


Figure 5. The normalized PL spectra measured at 15 K with 70 mW excitation power.

electronic coupling between QDs in vertical direction [13]. In addition, we measured the samples at three more different excitation power values (20 mW, 35 mW, and 44 mW) and compared their normalized PL spectra to observe power-dependent behavior (Fig. 6). Unlike in the case of single QD layer sample (SG), only the vertically coupled QD samples show the clearly observable amount of red-shift in the peak positions with growing excitation power. This phenomenon is regarded as the enhanced band-gap renormalization effect due to the increase of effective confinement volume arised from the vertical

coupling between QDs [14].

We applied multi-peak Gaussian fitting to analyze power dependency in PL spectra, and the fitting results for the spectra measured at 70 mW are presented here (Fig. 7). To start, the spectrum of a single QD layer (SG) can be fitted with two Gaussian peaks which correspond to the ground state (one peak at the lower energy side) and the first excited state (the other peak at the higher energy side). The shapes of PL spectra from this sample (SG) were almost unchanged with power variation, except the small rising of a tail at higher energy side originated from the more pronounced excited state peak. On the other hand, the samples of vertically coupled QD layers have the best fitting results with three Gaussian peaks. While the SL thickness was reduced from 10 nm to 8 nm, and then to 5 nm, the energy difference between the two most intense peaks in each spectrum was increased as 50.5 meV, 61.6 meV, and 67.9 meV, respectively. This is considered to be caused by material intermixing effect due to the proximity of two QD layers. As the SL thickness reduces, the strain originated from the first QD layer at bottom is able to affect the formation of the second QD layer at top more intensely. This effect may induce different ternary material compositions of QDs which renders the QDs to emit at higher energies [15,16].

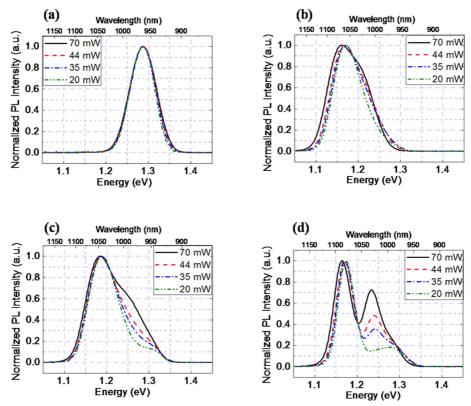


Figure 6. The normalized PL spectra measured at 15 K with different excitation power values from 20 mW to 70 mW: (a) SG, (b) SL 10 nm, (c) SL 8 nm, and (d) SL 5 nm.

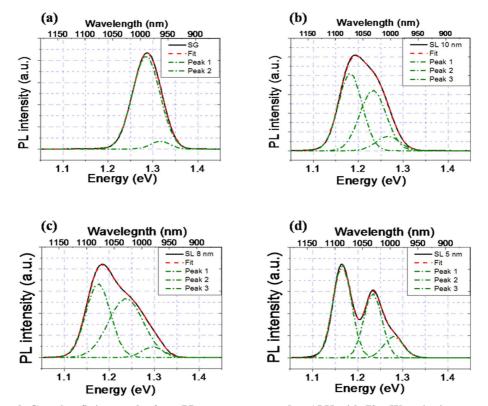


Figure 7. Multi-peak Gaussian fitting results from PL spectra measured at 15 K with 70 mW excitation power: (a) SG, (b) SL 10 nm, (c) SL 8 nm, and (d) SL 5 nm.

IV. Conclusions

We conducted excitation power-dependent PL measurements

at cryogenic temperature (15 K) to characterize the vertically coupled low density InGaAs/GaAs QDs grown by MEMBE method. The low surface density of QDs (~5 µm⁻²) and

vertical coupling between QDs were successfully observed by atomic force microscopy and scanning transmission electron microscopy, respectively. Our analysis on PL spectra identified the enhancement in the degree of vertical coupling between QDs according to the reduction of SL thickness from 10 nm to 5 nm. We attribute this pronounced modification of QD emission properties to the mini-band formation, band-gap renormalization, and material intermixing effects. These characterization results would contribute to the utilization of this material system towards implementation of quantum information processing devices.

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References

[1] S. Maier, P. Gold, A. Forchel, N. Gregersen, J. Mørk, S. Höfling, C. Schneider, and M. Kamp, Opt. Expr. 22, 8136 (2014).

- [2] M. J. Conterio, N. Sköld, D. J. P. Ellis, I. Farrer, D. A. Ritchie, and A. J. Shields, Appl. Phys. Lett. 103, 162108 (2013).
- [3] E. Koroknay, W. M. Schulz, D. Richter, U. Rengstl, M. Reischle, M. Bommer, C. A. Kessler, R. Roßbach, H. Schweizer, and M. Jetter, Phys. Status Solidi B 249, 737 (2012).
- [4] A. Rantamäki, G. S. Sokolovskii, S. A. Blokhin, V. V. Dudelev, K. K. Soboleva, M. A. Bobrov, A. G. Kuzmenkov, A. P. Vasil'ev, A. G. Gladyshev, and N. A. Maleev, Opt. Lett. 40, 3400 (2015).
- [5] R. Oulton, A. I. Tartakovskii, A. Ebbens, J. J. Finley, D. J. Mowbray, M. S. Skolnick, and M. Hopkinson, Physica E 26, 302
- [6] M. Scholz, S. BŠettner, O. Benson, A. I. Toropov, A. K. Bakarov, A. K. Kalagin, A. Lochmann, E. Stock, O. Schulz, and F. Hopfer, Opt. Expr. 15, 9107 (2007).
- [7] J. Claudon, J. Bleuse, N. S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne, and J.-M. Gérard, Nature Photon. 4, 174 (2010).
- [8] N. K. Cho, S. P. Ryu, J. D. Song, W. J. Choi, J. I. Lee, and H. Jeon, Appl. Phys. Lett. 88, 133104 (2006).
- [9] J. D. Song, Y. J. Park, I. K. Han, W. J. Choi, W. J. Cho, J. I. Lee, Y. H. Cho, and J. Y. Lee, Physica E 26, 86 (2005).
- [10] S.-P. Ryu, N.-K. Cho, J.-Y. Lim, A.-R. Rim, W.-J. Choi, J.-D. Song, J.-I. Lee, and Y.-T. Lee, Jpn. J. Appl. Phys. 48, 091103 (2009).
- [11] J. S. Wang, S. H. Yu, Y. R. Lin, H. H. Lin, C. S. Yang, T. T. Chen, Y. F. Chen, G. W. Shu, J. L. Shen, and R. S. Hsiao, Nanotechnology 18, 015401 (2007).
- [12] M. A. Migliorato, L. R. Wilson, D. J. Mowbray, M. S. Skolnick, M. Al-Khafaji, A. G. Cullis, and M. Hopkinson, J. Appl. Phys. 90, 6374 (2001).
- [13] G. S. Solomon, J. A. Trezza, A. F. Marshall, and J. S. Harris Jr, Phys. Rev. Lett. 76, 952 (1996)
- [14] J. Rihani, N. B. Sedrine, V. Sallet, M. Oueslati, and R. Chtourou, Appl. Surf. Sci. 254, 3125 (2008).
- [15] M. O. Lipinski, H. Schuler, O. G. Schmidt, K. Eberl, and N. Y. Jin-Phillipp, Appl. Phys. Lett. 77, 1789 (2000).
- [16] Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, Phys. Rev. Lett. 75, 2542 (1995).