Intermediate band formation by InGaAs/GaAs multistacked quantum dots without strain compensation

Keishiro Goshima¹, Keisuke Inukai¹, Norio Tsuda¹, and Takeyoshi Sugaya²

¹ Aichi Institute of Technology, 1247 Yachigusa Yakusa, Toyota, Aichi, Japan
² Advanced Industrial Science and Technology, 1-1-1 Umezono Tsukuba, Ibaraki, Japan

Abstract. Quantum dots (QDs) are widely used for enhancing the performance of optical devices. Intermediate-band solar cells that use multistacked QDs can surpass the conversion efficiency of conventional silicon cells. We performed the optical characterization of In₀.₄Ga₀.₆As/GaAs multistacked QDs without strain compensation. We used the theoretical and experimental techniques prescribed for intermediate-band solar cells, i.e., photoluminescence (PL) spectroscopy and two-color excitations spectroscopy. The interdot spacings were not uniform and were found to be 15 nm and 7 nm. The results verify the formation of intermediate bands by the multistacked QDs. Using the theoretical studies and experimental results, we performed an in-depth study on the mechanism underlying the formation of intermediate bands by the multistacked QDs and the effect of different interdot spacings.

1. Introduction

Recently, self-assembled quantum dots (QDs) have been shown to be possible candidates for solar cells, semiconductor lasers, and quantum cryptography mediums. To realize these optoelectronic devices, it is necessary to increase the optical gain in QDs. Multiple-stacking in QDs is one possible solution for this[1]. Moreover, multistacked QDs not only exhibit a higher optical gain but can also realize an intermediate band by controlling the barrier thickness between the QDs. For example, intermediate solar cells achieved by creating the intermediate band between the valence band and the conduction band using quantum mechanical coupling. Photo carriers can arise with the cascade process from the conduction band to the valence band via the intermediate band, and they are used to lower the energy photon below the band gap energy. Thus, the ultimate conversion efficiency at sun intensity can increase above 60%[2,3]. The intermediate band structure is realized by a superlattice structure grown by multistacked QDs. The intermediate band in the QD array is caused by the electronic coupling of states in the neighboring QDs. When the interdot spacing between the QDs of growth detection is less than the de Broglie length, the QD states eventually delocalize over the QDs arrays, such as the coupling effect, and new energy bands with the existing carriers are formed [4]. To realize intermediate solar cells, it is necessary to obtain multistacked QDs structures with small interdot spacings. Meanwhile, growth techniques and self-assembled QDs with multistacked growth have been studied. The crystal quality of QDs degrades owing to issues such as crystal defects and transitions in crystals resulting from the internal strain. Therefore, strain compensation techniques have been introduced to fabricate multistacked QDs with good crystal quality [1,5].

However, the disadvantage of this technique is that it cannot fabricate small interdot spacing structures. Multistacked QDs with an intermediate band require a small interdot spacing because they use the coupling effect owing to the quantum mechanical interactions. We studied the fabrication of InGaAs/GaAs QDs because InGaAs has a slight lattice mismatch with the GaAs substrate. We successfully fabricated small interdot multistacked InGaAs QD structures without
using a strain compensation technique by the intermittent deposition of InGaAs layers using an As$_2$ source [6,7]. In previous studies, we observed the electronic band structure and properties of multistacked QDs with different interdot spacings [8,9]. In this study, we investigated the intermediate-band (sub-band) formation and the cascade excitation process in multistacked QDs with various barrier thicknesses by theoretical analysis and optical measurements.

2. Sample and Experimental Procedure

The structure was fabricated by the intermittent deposition of In$_{0.4}$Ga$_{0.6}$As layers, using an As$_2$ source. Consequently, the crystal quality was not degraded. We grew ten-stack In$_{0.4}$Ga$_{0.6}$As QD layers on a Si-doped GaAs(001) substrate using an As$_2$ source by molecular beam epitaxy. Figure 1 shows the schematic of the multistack QD structure. We grew ten-stack In$_{0.4}$Ga$_{0.6}$As QD structures with a GaAs barrier layer of the thicknesses 10 nm and 20 nm, corresponding to the interdot spacing of 7 nm and 15 nm, respectively. The height, width, and shape of a dot were 5 nm, 20 nm, and pyramidal, respectively. Further details of our growth process can be found in the literature [6,7]. Photoluminescence (PL) was investigated using a spectroscopy system. Laser light was focused on the QD samples through objective lens. Pulse laser with the pulse width of 150 ps and the repetition rate of 50 MHz was used as an excitation source in the PL measurements. The QD sample was mounted on a helium cryostat, and the temperature was tuned to 10 K. The PL signals were detected by a monochromer, whose spectrum resolution was 0.1 nm, and a silicon-avalanche photodiode (Si-APD) detector was used. We used two color lasers: color 1 laser was a pulse laser with a wavelength of 780 nm, and color 2 laser had CW operation with a wavelength of 1300 nm. The energy of color 2 laser tuned to the ground state of the QDs.

3. Results and Discussion

3.1 Numerical Circulation

Figure 3 shows the numerical calculations of the electric structures according to barrier thickness. To compute the electronic states of the QD embedded in the GaAs surrounding, we solved the Schrödinger equation in the effective mass approximation using three-dimensional finite element modeling using COMSOL Multiphysics [10]. We calculated that the bonding and anti-bonding states for the electron were formed by a barrier with a thickness less than 10 nm. The lesser the barrier thickness, the more the energy deference between the bonding and the anti-bonding states were spread. In another case, the bonding and anti-bonding states for the hole were formed within 4 nm. These different results were caused by the effective mass between the electron and hole. We expected perfect intermediate-band (sub-band) states to be formed by a barrier thickness of within 4 nm.
3.2 Photoluminescence

We observed the PL at 10 K. Figure 4 and 5 show the PL spectra for the interdot spacings of d = 15 nm and d = 7 nm, respectively. For d = 15 nm, the primary peak and FWHM of the PL spectrum were 1.25 eV and 53 meV, respectively. From the calculation results, the wavefunction was not coupled, and the intermediate band was not created because of the large interdot spacing. Therefore, the spread of the PL spectra was caused by the size deviation of the individual QDs.

Meanwhile, for d = 7 nm, three peak components are shown in the PL spectrum. The primary peak and two subpeaks were 1.29 eV, 1.317 eV, and 1.253 eV, in which the peaks were identified by fitting a Gaussian curve. From the calculation results, the electron wavefunction was coupled, and the intermediate band was created because of the small interdot spacing. We now discuss the primary peak of 1.29 eV, and the subpeak of 1.317 eV. It was assumed that the primary peak and subpeak energies were caused by the transition regime from the bonding and anti-bonding states of the electron, respectively. The observed energy difference between the primary and subpeaks was 15 meV. The FWHM of d = 7 is 65 meV, which is larger than d = 15 nm. The difference of FWHM between two samples was 12 meV, which is expected the energy difference between the bonding and anti-bonding states with d = 7 nm. This result may in fact show a coupling effect about d = 10 nm or less. The peak of 1.253 eV is caused by the enlargement of QDs in the stacking direction because of the lower energy side of the primary peak [11].

![Figure 4: PL spectrum of the d = 15 nm sample at 10 K. Dashed line shows the three components used for fitting.](image)

3.3 Two-color excitation photoluminescence

To certify the intermediate band, we performed the carrier dynamics using two-color excitation. Figures 7 and 8 show the photoluminescence spectra with ground state in the multistacked QDs. The interdot spacing of these sample were d = 15 and d = 7 nm, respectively. We used two color lasers: color 1 laser was a pulse laser with a wavelength of 780 nm, and color 2 laser had a CW operation with a wavelength of 1300 nm. One-color (only color 1) and two-color (color 1+2) excitations are shown with black and red lines, respectively.

In the case of d = 15 nm, the PL intensities did not change between one-color and two-color excitations. However, in the case of d = 7 nm, we observed that the PL intensities were lower for the two-color excitation. Figure 2 shows the band diagram of the multistacked QDs with an intermediate band. In the one-color excitation, the carriers were initially excited directly to the conduction band by the first laser. Subsequently, the excited carriers relaxed to the ground state in the QDs. Finally, the carriers recombined with the PL to the hole ground state. In the two-color excitation, the second laser excitation energy was tuned below the ground-state energies. The photocarriers excited by the first laser relaxed to the intermediate state in the QDs. However, these relaxed carriers were excited again from the intermediate band to the conduction band by the second laser, as follows in Fig.8.9. Therefore, we assumed that the recombination from the ground state decreased by the cascade process into the carriers via the intermediate band. In addition, the peak and FWHM of the PL spectra at d = 7 nm did not change...
under one-color and two-color excitations. We assumed that the carriers in intermediate band were uniformly re-excited by the second excitation.

4. Summary

We investigated the electronic structures, intermediate formation, bandwidth, and carrier dynamics for different interdot spacings of multistacked QDs using optical methods. From the PL spectra method, we observed that the shape of the spectra was different for interdot spacings of $d = 15$ nm and $d = 7$ nm. We assumed that the broadening of the spectra for $d = 7$ nm was caused by the anti-bonding and bonding states of the electron, and the enlarged QDs in the stacking direction. For the one-color and two-color excitation methods, we observed different PL intensities for different interdot spacings. The intensity of the PL peak decreased for $d = 7$ nm when the two-color excitation was used. This behavior was caused by the carrier dynamics that reactivated the process via the intermediate state.

References

[1] Akahane K, Ohtani N, Okada Y and Kawabe M 2002 J. Cryst. Growth 245 31-36
[2] Luque A and Marti A 1997 Phy. Rev. Lett. 78(26) 5014-5017
[3] Antolion E, Marti A, Farmer C D, et al. 2010 J. Appl. Phys. 108 064513
[4] Tomic S 2010 Phys. Rev. B. 82 195321
[5] Jang Y D, Badcock T J, Mowbray D J, Skolnick M S, et al. 2008 Appl. Phys. Lett. 92 251905
[6] Sugaya T et al. 2011 Solar Ener. Mater. Solar Cells 95 2920-2923
[7] Sugaya T et al. 2010 Jpn. J. Appl. Phys. 49 030211
[8] Goshima K, Tsuda N and Sugaya T 2018 Jpn. J. Appl. Phys. 57 06HE08
[9] Goshima K, Tsuda N and Sugaya T 2016 Photovoltaic Science and Engineering Conference.1-1-1e
[10] Melnik R and Willatzen M 2004 Nanotechnol. 15 1-8
[11] Wasilewski Z R et al. 1999 J. Cry. Grow. 201-202 1131-1135