# Report on AOARD-06-4017
(Contract No. :FA5209-06-P-0183 AOARD 06-17)

## *Summary of the proposal*

Title : “Growth and Characterization of low density In(Ga)As/GaAs quantum dots for quantum information processes”

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Final goal : Growth of low density QD substrate for quantum information processes
1st year’s goal (2006): Growth of In(Ga)As/GaAs QD and its characterization.
(QD density ~ $1/\mu m^2$ , μ-PL measurement setup for single spectroscopy)
Approach ;

- Study on growing parameters for low density In(Ga)As/GaAs QDs with migration enhanced molecular beam epitaxy (MEMBE) and growth of low density QDs.
- Characterization of wetting layer in low density QDs with a macro-PL measurement.
- Development of μ-PL measurement for a single QD spectroscopy at low temperature ~ 4 K.

## *Report of the project*

### I. Introduction of the project

Semiconductor single quantum dot (QD) offers many potential applications in quantum information processes such as an all-optical quantum gate1), a single-photon source for the quantum cryptography2) and entangled states3) for the quantum computation. For such applications, low-density QDs are desirable for an easy manipulation of them through standard semiconductor device fabrication processes and test processes, since many QDs in a micron-sizes (or wavelength-sized) area can interfere each other to avoid single QD-based functionality.

Almost all InAs/GaAs QDs reported to date have been grown by Stranski-Krastanov (SK) mode, in which InAs is supplied continuously. Allooing et al. 2) have reported QD density of 2 dots/$\mu m^2$ by using very low growth rate of InAs ( ~ 0.0012 mono-layer (ML)/s) and a successful demonstration of spectroscopy of a single QD. In our approach to grow low density InAs/GaAs QDs, we have used alternating supplies of In and As using migration enhanced epitaxy. We can

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14. ABSTRACT
**The work demonstrated growth of low density QDs and optimized of growth parameters for low density In(Ga)As/GaAs QDs with enhanced molecular beam epitaxy (MEMBE). The work also included characterization of wetting layer in low density QDs with a macro-PL measurement and development of &#61549;-PL measurement for a single QD spectroscopy at low temperature ~ 4 K.**

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control the size and density of QDs by controlling the interruption time between the supply of In and As.4) In this project, we tried to develop growing technology of low density In(Ga)As/GaAs QDs (~ 1 dots/μm$^2$ ) for real applications. We also tried to investigate the spectroscopic behavior of a single QD to understand the nature of it by using CL and micro-PL measurements.

## II. Growth of low density In(Ga)As/GaAs QDs with MEMBE (Migration Enhanced Molecular Beam Epitaxy) technique

In(Ga)As/GaAs QDs have been grown through the mechanisms releasing the strains accumulated in In(Ga)As layer on GaAs due to a lattice mismatch ( ~ 7%) to form 3-dimensional structures (i.e. QDs) when the supplied In(Ga)As layer is thicker than the critical thickness. In this conventional method (Stranski-Krastanov (SK) method), In, (Ga), As are simultaneously supplied to grow In(Ga)As/GaAs QDs. This method accompanies the relatively thick 2-dimensional structure (wetting layer). However, when we introduce the time intervals (growth interruption) between the deposition of group III atoms, such as In and/or Ga, and the deposition of As, we can control the migration of In and/or Ga on the surface of GaAs to form a 3-dimensional structure. This kind of migration enhanced epitaxy (MEE) technique has been reported to provide InAs/GaAs QDs with a better uniformity with larger sizes and thinner wetting layer which showed temperature-insensitive PL (photoluminescence) linewidth compared to the SK technique. 4) One can control the size and composition of QDs by controlling the periods of each layer, the content of materials in the layer and growth interruption (GI) time. Figure 1 shows schematics of the growth of InAs/GaAs QDs with GI time between the deposition of In and As. This technique has one more growing parameter, GI time to grow QDs, which enable us to control the size of QDs.

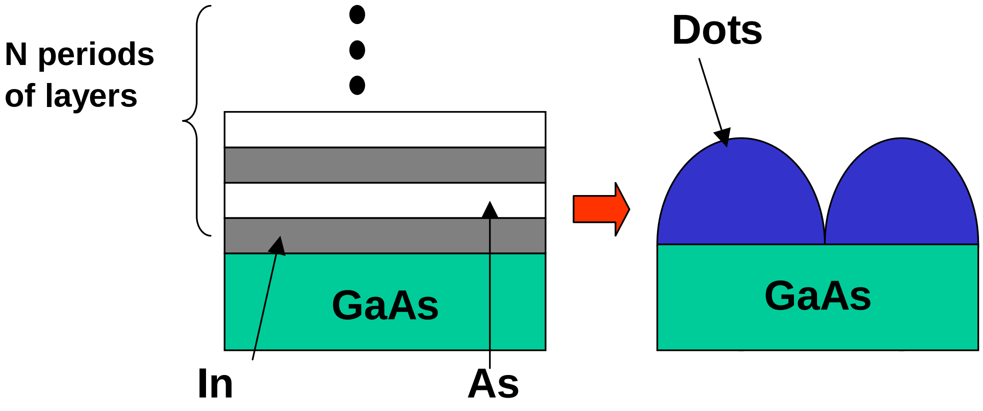

Fig. 1 Schematics for the growth of InAs QDs with MEE technique

The growing parameters for In(Ga)As/GaAs QDs with MEMBE method are the fluxes of elements (In, Ga, and As), substrate temperature, and GI time. Figure 2 shows QD density as a function of GI time with fixed substrate temperature and the fluxes of In and As. The inset shows AFM (Atomic force Microscope) images of QDs at each GI time. As one can see in this figure, the dot density decreases with increasing GI time. At shorter GI time, there are many small dots with larger dots. But as increasing the GI time, the number of small dots decreases and the dot size increases, since the small dots (i.e. In atoms in small dots) can move to make larger dots during GI time.

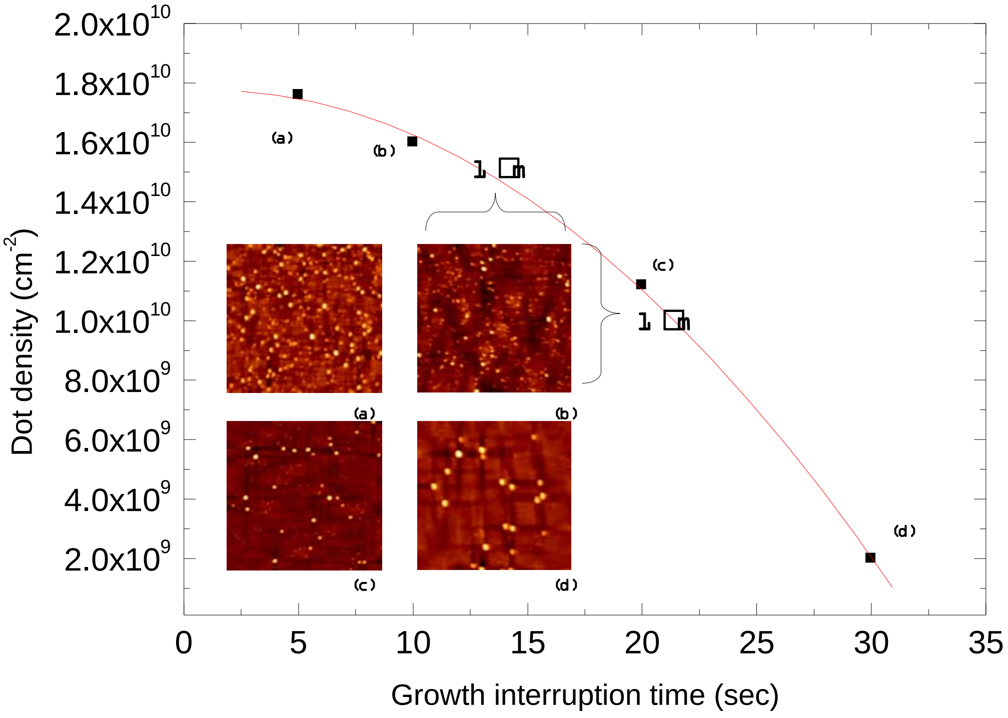

Fig. 2. Density of InAs/GaAs QDs as a function of GI time

Figure 3 shows dot density as a function of As-flux which is determined by the temperature of Asenic cell in MBE machine. In the experiment, GI time was fixed at 30 sec. One can easily see that small amount of As-flux (i.e. at low As cell temperature) provides low dot density with fixed In-fulx. This implies that the migration length of In atoms are relatively short with large amount of As-flux, which makes In atoms to be easily fixed and makes high dot density. One can see that there are no small dots in AFM image (b) in Fig. 3, for large amount of As-flux, which means the aggregation of small dots to larger dots. So one can conclude that low As-flux is essential to grow low density InAs/GaAs QDs.

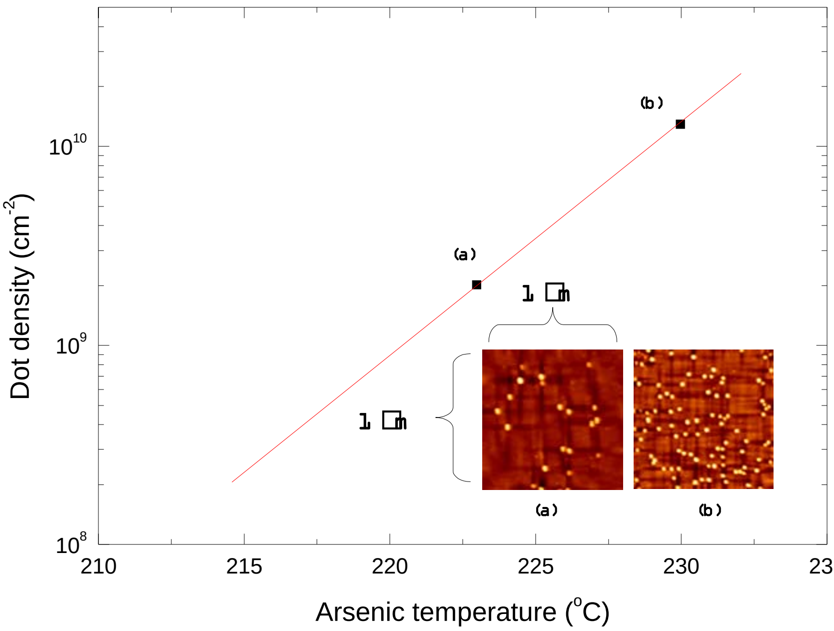

Fig. 3. Density of InAs/GaAs QDs as a function of As-flux (As-cell temperature)

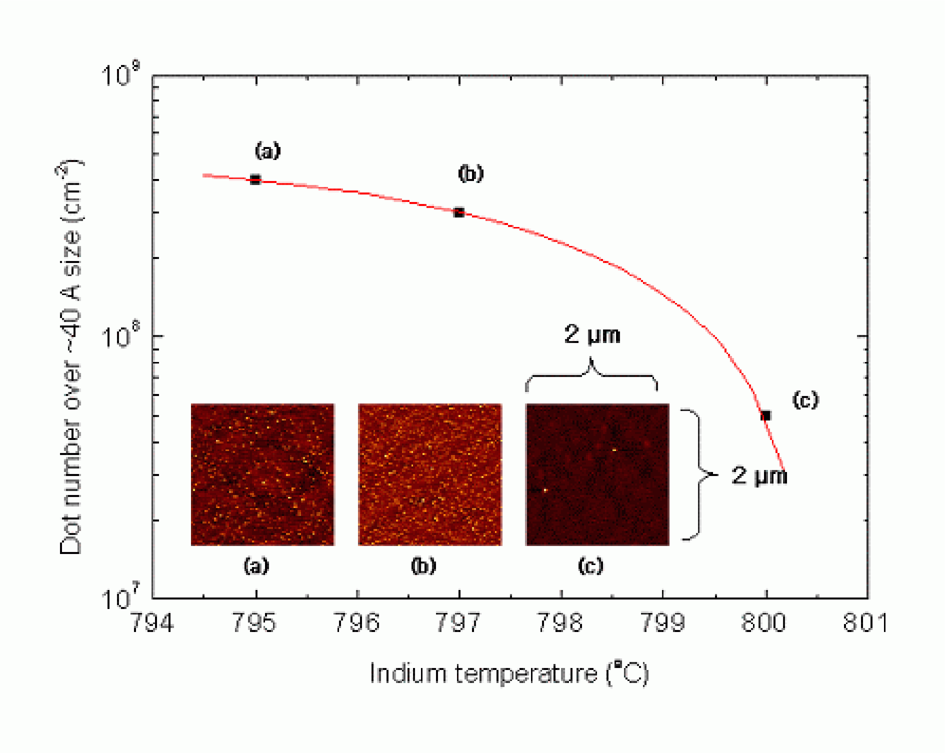

Fig. 4. Density of InAs/GaAs QDs as a function of In-flux (In-cell temperature)

Figure 4 shows dot density as a function of In-flux which is determined by the temperature of Indium cell in MBE machine. In the experiment, As-flux was set to be low in order to get high In migration length. In this figure, one can see the decrease of dot density and the increase of dot size with increasing In-flux. We could get low density ( ~ 2 QDs / $\mu m^2$) InAs/GaAs QDs at high in-cell temperature (i.e. high In-flux) with low As pressure. In atoms in the layer under high In-flux with low As-flux cannot be used to make QDs completely, remain in metal state to be evaperated during GI time.

Physicists need larger QDs because large QD can provide large oscillator strength and large overlap of cavity mode and single QD in a micro-cavity, which is good for the study of semicondutor-based quantum optics or quantum electro-dynamics. Since the large oscillator strength of a electron-hole pair (i.e. exciton) induces short decay time, the larger QDs can make single photon trains with a short pulse cycle, which is good for a single photon source. The short decay time of large QD is thought to be good for the electrically-driven single photon source which does not use micro-cavity. When large QD is adapted in a micro-cavity, the overlap of an optical cavity-mode and the QD increases. In this situation, the quantum mechanical system, QD in a micro-cavity, can be easily modified with few photons to be absorbed by the QD in its quantum mechanical characteristics. This kind of quantum mechanical system with large QD in a micro-cavity is better suited for the study of quantum information processes, such as quantum logic gates and quantum computing based on quantum optics. Furthermore, large semiconductor QDs can have many quantum states (s-, p-, d-, and f-state), depending on its size, to be studied basically for real applications. For practical experiments, physicists also need low density QDs with their emitting wavelength shorter than 1 μm because they want use the most sensitive LN2 cooled CCD-array due to the difficulties in detecting few photons from a single QD in a wide wavelength range for the basic study of QD in a micro-cavity, even though QDs should emit 1300 nm or 1500 nm of wavelength for a quantum cryptography at the optical communication band. Since the larger size of QD induces the longer wavelength, it is impossible to grow InAs/GaAs QDs for such requirement. Though InGaAs QD has been grown for the above requirement, it's not simple to match the requirement. In this project, we tried to grow low density InGaAs/GaAs QDs with large size by using MEMBE technique.

Figure 5 shows AFM image of low density (~ 5QDs/$\mu m^2$) InGaAs/GaAs QDs grown by MEMBE method. The width and height of QD are about 75 nm and 5 nm, respectively. Since the QD size is larger, one can expect the larger oscillator strength, shorter decay of excitons bounded in the QD and large nonlinear properties with it in the high-Q cavity such as PC-based cavity and DBR-based cavity. The AFM image shows 5 different sized QDs in a 1x1 $\mu m^2$, which means broad inhomogeneous broadening in its PL spectrum. The density of 5 QDs/$\mu m^2$ is enough for a single-dot spectroscopy, since one can isolate the signal from a single QD spectroscopically. The InGaAs QDs in a GaAs matrix shows the ground state peak at 980 nm with a broad inhomogeneous broadening in Fig. 6. The power dependent macro-PL measurement (Figure 6) shows ground (GS) state at 980 nm and 1st excited state (ES) at 945 nm. Since the GS spectrum is wide, one can expect that the number of QDs emitting shorter wavelength than 980 nm is small, which is good condition to select QDs emitting shorter wavelength to study single dot spectroscopy.

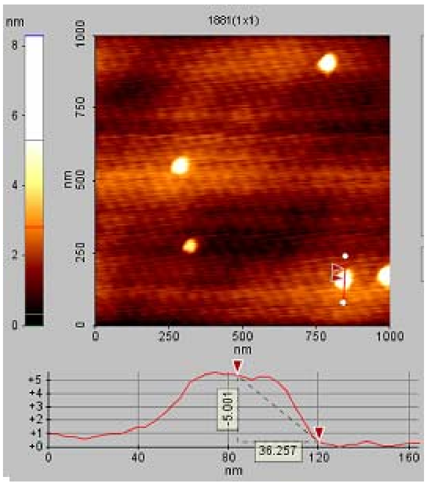

Fig. 5 AFM image of InGaAs/GaAs QDs. The size of QD is 5x75 nm$^2$.

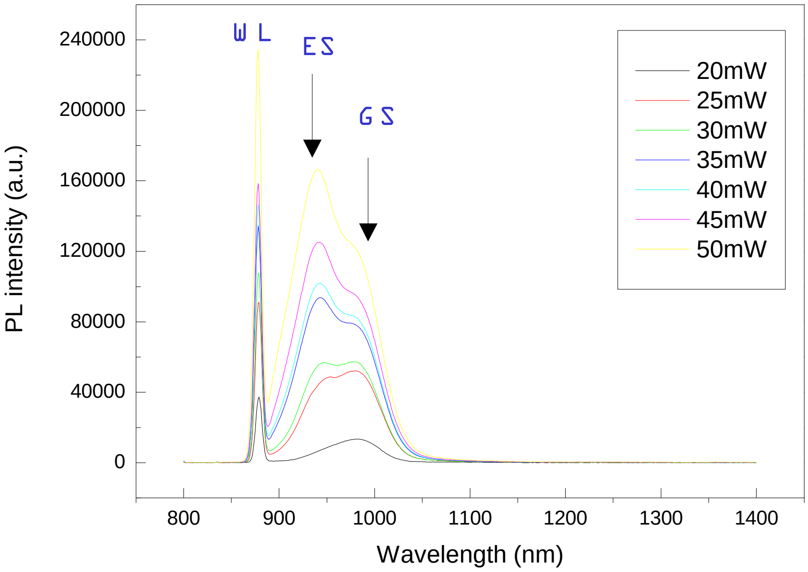

Fig. 6. Power-dependent macro-PL spectra measured at 10 K. GS: ground state, ES: excited state, WL: wetting layer.

## III. Characterization of low density In(Ga)As/GaAs QDs by spectroscopic techniques

Spatially resolved CL measurement of the sample described in Fig. 6 was carried out at 5K along the masking edge of sample holder. The CL spectra showed clear luminescence peaks from several confined states in QDs. The dot density was varied with the position. Figure 7 shows CL spectra measured near the masked region due to the sample holder for the growth. Few CL peaks can be found only inside the masked region.

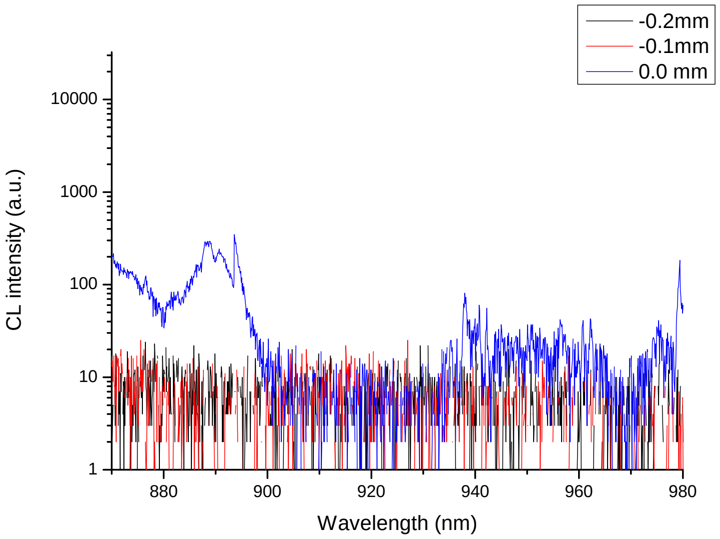

Fig. 7. CL spectra taken near masking edge

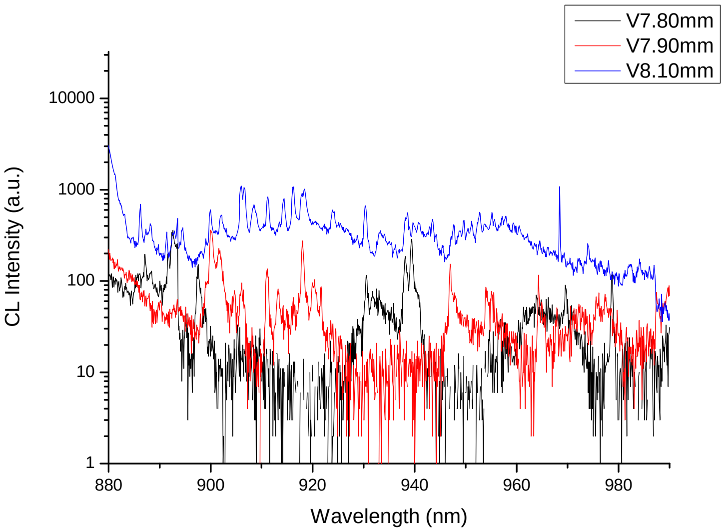

Fig. 8. CL spectra taken the positions apart from masking edge by 0.1, 0.2, and 0.4 mm.

Figure 8 shows CL spectra taken at the positions apart from the edge of masked region by 100 μm ~ 400 μm. There are increased single QD peaks with the position far apart from the masked region. In this range, many QD lines are located at shorter wavelength, which means that the QD sizes are small or the QDs have a high Ga mole fraction.

Figure 9 shows CL spectra taken at the positions apart from the edge of masked region by 600 μm ~ 1200 μm. One can see the decrease of CL intensity to the position of 1.0 mm. The larger background intensity of CL signal at those regions may be due to the stronger effect of wetting layer. The background intensities of CL signal at 1.0 mm and 1.2 mm are same, which means the dot densities are similar in this region. As one can see in Fig. 7 and Fig. 9, the relative CL intensity at longer wavelength increases with the position closer to the center of the sample. This result implies that the size of QDs becomes larger at the positions far apart from the masked region. Therefore one can conclude that the sample has a uniform dot density with large sizes inside the region apart from the masked region by 1.0 mm.

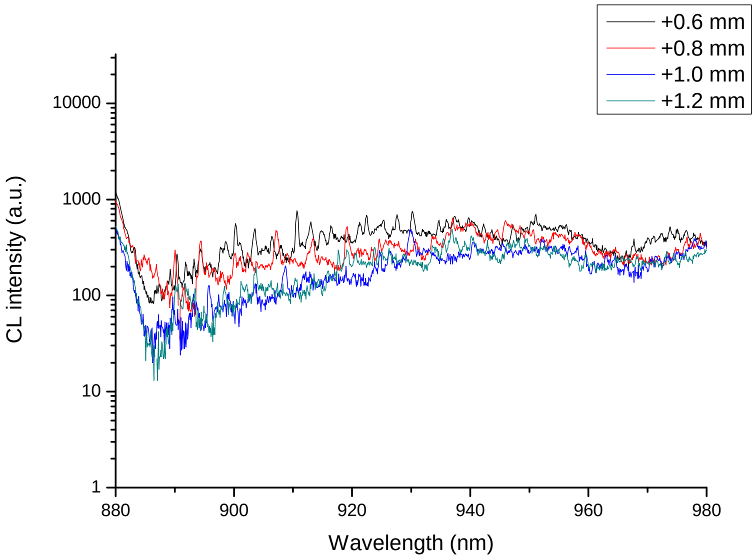

Fig. 9. CL spectra taken the positions apart from masking edge by 0.6, 0.8, 1.0, and 1.2 mm.

In order to measure the spectrum from a single QD, micron-sized mesa was patterned. μ-PL measurement at 6K for a mesa (1x1 μm$^2$) showed many peaks from the confined excitons in each QD of the mesa as in Fig. 10. Since there are many GS signals (exciton, bi-exciton etc) and ES signals from QDs in 1x1 μm$^2$ sized mesa, it is difficult to determine which signals come from a specific single QD. Since ES state signals cannot be detected at low pumping condition, one can identify the signal from the GS state of QDs. Figure 11 shows power dependent micro-PL spectra. There is only one PL peak at 912.3 nm at low pumping condition. But its intensity increases with another peaks at 913.1 nm as the pumping power increases. Therefore one can conclude that the peaks at 912.3 nm and 913. 1 nm come from exiton and bi-exciton from the same QD. One can also see the exciton and bi-exciton signal from another QD near 918 nm.

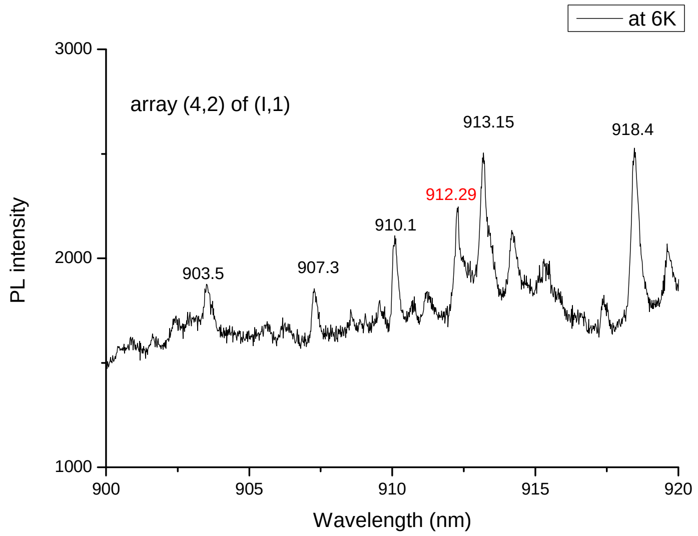

Fig. 10. PL spectra measured at 6 K taken from 1 μm$^2$ sized mesa in the wavelength range from 900 nm to 920 nm.

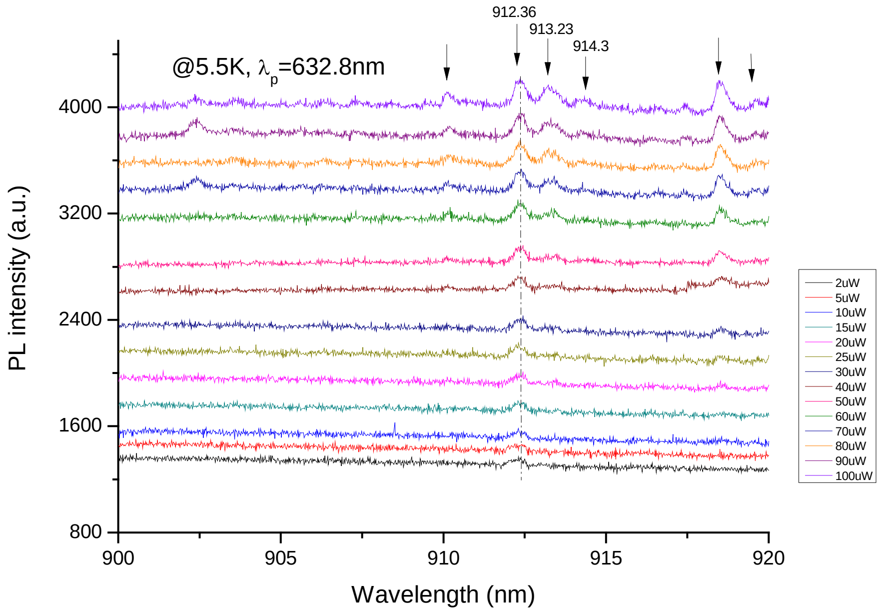

Fig. 11. Power dependent PL spectra at 5.5 K taken from 1 μm$^2$ sized mesa in the wavelength range from 900 nm to 920 nm.

Temperature dependent PL measurement was also carried out for the same mesa in order to characterize the temperature dependent behavior of exciton and bi-exciton from large QD. As one can see in Fig. 12, the peak at 912.3 nm ( i.e. exciton peak) still alive up to 40 K without any change in its emission wavelength. This result is particularly interesting for the design of temperature-insensitive single photon source.

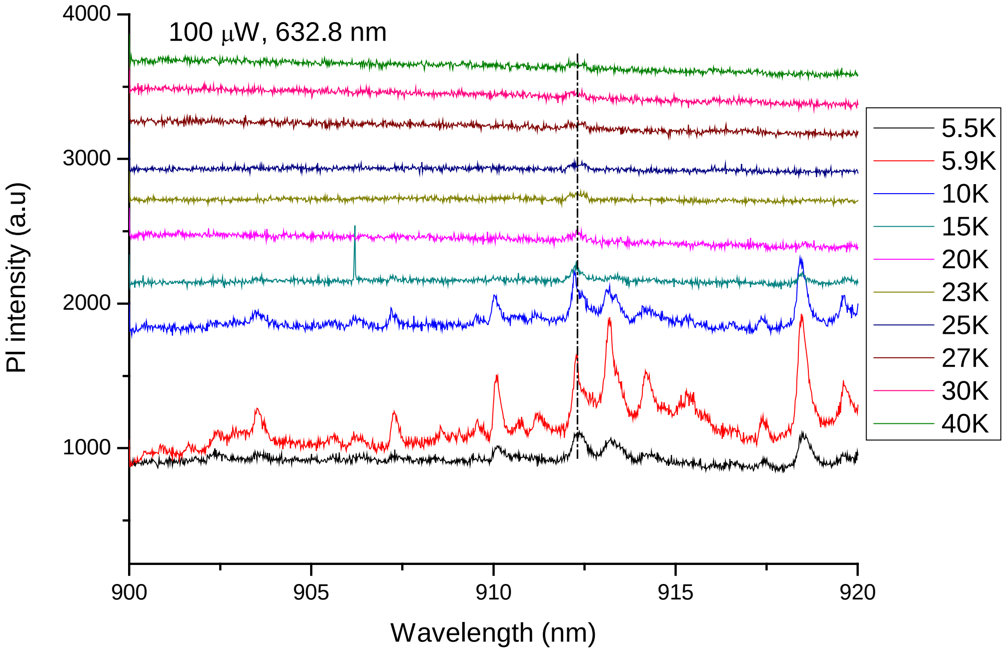

Fig. 12. Temperature dependent PL spectra at 5.5 K taken from 1 μm$^2$ sized mesa in the wavelength range from 900 nm to 920 nm with pumping power of 100 μW.

## IV. Conclusion and Future works

We have grown low-density InAs/GaAs (~0.5 QDs/μm$^2$ ) and InGaAs/GaAs (~5 QDs/μm$^2$ ) quantum dots (QDs) by using migration enhanced molecular beam epitaxy (MEMBE), without any rotation stop during the growth of QDs. The spatially resolved CL measurements at 5K showed that the density of InGaAs QDs is uniform inside regions apart by 1 mm from the substrate holder. The use of MEMBE for the growth of low-density QDs is well adapted to the large-area fabrication of single-dot based optical devices, such as single photon sources and quantum logic gates. The height and width of InGaAs QDs are 5 and 72 nm, respectively. Their emission band at 10 K exhibits two peaks at 980-nm and 940-nm corresponding to the ground state and the 1st excited state transitions, respectively. The large QDs are thought to be better suited for the study of quantum electrodynamics and quantum logic gate utilizing a single QD in micro-cavity, since they have larger oscillator strength as well as larger coupling with the optical cavity mode. Single QD spectroscopy for InGaAs QDs at low temperature was performed using a mesa-patterned sample and a flow-type liquid He cryostat. The exciton and bi-exciton peaks were identified by using power-dependent micro-PL measurements. We have observed the exciton peak up to 40 K without any change in its emission wavelength. This is particularly interesting for the design of temperature-insensitive single photon source.

Since large QDs are known to have large oscillator strength and short decay time, we need to measure the decay time of exciton of it and compare it with the result from small QDs. The single QD in a micro-cavity is interesting topic both for basic understanding of quantum mechanical nature of it and for the real application of it to a single photon source. We will tried to make a single QD in photonic-crystal based micro-cavity and characterize it.

## References

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