

Low-Temperature Micro-PL Measurements of InAs Binary Quantum Dots on GaAs Substrate

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ABSTRACT

Optical properties of InAs binary quantum dot (bi-QD) molecules grown on the (001) GaAs substrate were measured by means of temperature- and excitation-power-dependent photoluminescence (PL) spectroscopy. It was observed that the shape and peak position of the PL spectra changed with the temperature and with the excitation power. It was also found that the linear polarization degree of the bi-QD PL signal changed with temperature. The temperature-dependent PL described that the linear polarization degree of bi-QDs is closely related to the carrier dynamics.

Keywords: binary quantum dot, excitation-power-dependent photoluminescence, temperature-dependent polarization, polarization anisotropy, carrier dynamics

1. INTRODUCTION

Quantum dots (QDs) are promising candidates as the building block of future optoelectronic devices and quantum information technology [1-3]. Since the charge carriers in quantum dots are confined in all three spatial directions, this leads to a discrete energy spectrum. Because of this behavior, semiconductor quantum dots have been expected to bring a significant progress in optical device applications. Many research groups have reported many quantum-dot structures, and this has attracted attention as possible implementation of future quantum nano-devices such as quantum-dot molecules for quantum cellular automata, etc. [4-5] Several techniques have been

proposed and investigated for the fabrication of quantum dots; among these the most promising method is that of self-organized growth. To understand optical properties of these semiconductor quantum-dot structures, photoluminescence (PL) spectroscopy is the most widely used optical characterization tool. Information on the PL of quantum dots is an important basis for the future development of novel nano-scale devices such as optical detectors, emitters, modulators, and lasers. The main idea of PL spectroscopy is investigating the transition of photo-generated carriers between the valence-band and conduction-band states. The results reflect the conduction and balance band structures, the dynamics of the relaxation and recombination, the excitonic and the many-body effects, etc. [6] However, the PL spectrum, which gives a number of such important information, is sometimes difficult to extract.

Recently, the formation of binary-quantum-dot (bi-QD) molecules was achieved on the GaAs (001) substrate by gas-source molecular-beam epitaxy (GSMBE) under arsenic (As₂) over pressure using a thin-capping and re-growth technique [7]. In this paper, a study of optical properties of these bi-QD molecules by means of micro-PL spectroscopy is presented, with a focus on the temperature-dependent and excitation-power-dependent PL, which can be monitored under non-resonant excitation. It was found that the optical polarization anisotropy of the PL signal arises from the temperature-dependent carrier dynamics.

2. SAMPLE PREPARATION

The bi-QD sample investigated in this work was fabricated and some preliminary results of which were reported in Ref. [7]. The QD density on the bi-QD sample is $1.1 \times 10^{10} \text{ cm}^{-2}$. All the bi-QD molecules on the sample are oriented along the crystallographic direction. The average center-to-center separation is 22 nm. An AFM image and a sketch of the bi-QDs are shown in Fig. 1 (a) and (b), respectively. The detail of the fabrication process can be found in Ref. [7].

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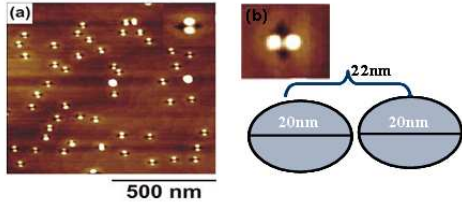


Fig.1: (a) An AFM image of the bi-QDs, and (b) a close-up AFM image of a bi-QD with dot size and separation indicated.

3. EXPERIMENTAL SETUP

All the new PL measurements were carried out with a standard micro-PL setup. The sample was mounted on the cold finger of a sample holder, which was cooled down by cold helium gas in a flow-type cryostat to between 3.8 K and 160 K. For micro-PL measurements, a cw: semiconductor laser (660 nm) was used as the excitation source. The excitation laser was focused to a 1-2 μm spot on the sample by a microscope objective lens, whose numerical aperture and working distance were 0.42 mm and 200 mm, respectively. The laser beam shape, size, and location on the sample were checked by monitoring the reflection image with a CCD camera, on which the laser beam was illuminated together with a white light illumination by a tungsten lamp. The luminescence of the sample was collected by the same objective lens, reflected off the mirror and passed through a band-pass filter to remove the excitation light. The image was then focused onto the entrance slit of a spectrometer and detected through with a liquid nitrogen-cooled InGaAs detector. For the polarization analysis, a rotating half-wave plate and a fixed polarizer were inserted in front of the spectrometer. The sketch of the experimental setup is shown in Fig. 2.

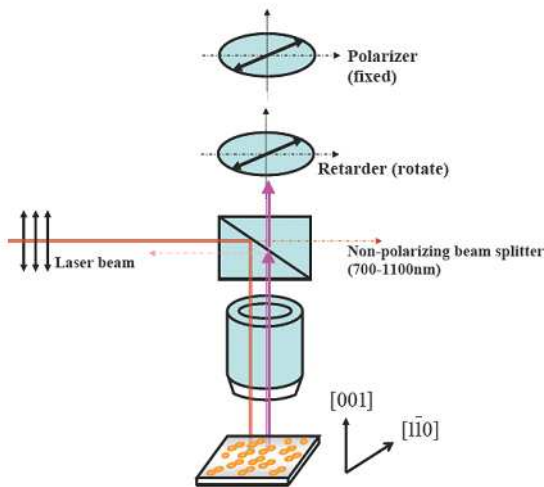


Fig.2: Experimental setup of the measurement.

4. RESULTS AND DISCUSSION

The bi-QD sample gave very strong PL at the peak emission wavelength of 1100 nm for excitation above the GaAs barrier band gap at 10 K. Room temperature PL measurements were then carried out. Fig. 3 shows various PL spectra measured at room temperature by using the excitation power ranging from 280 μW to 800 μW . The PL peak at 1100 nm (1.15 eV) was observed and the second peak around 953 nm (1.3 eV) distinctly occurred. The energy difference between first peak and second peak of about 145 meV is large enough that it can be then distinguished as the excitation state compared with typically 50-60 meV for isolated InAs quantum dots [8]. The second peak at 1.3 eV is attributed to come from the wetting layer of InAs quantum dots due to the low density of QDs on the sample. A full-width-at-half-maximum (FWHM) of around 70 meV of the first PL peaks obtained from a room-temperature measurement indicates the size fluctuation and thermal relaxation of carriers of QDs at high temperatures.

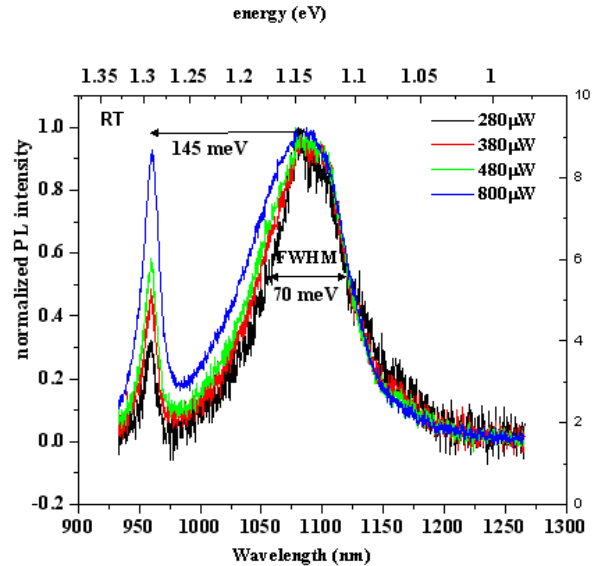


Fig.3: Room-temperature PL spectra obtained by using excitation power ranging from 280 μW to 800 μW .

Low-temperature PL measurements were also carried out to understand the physics of the bi-QD structure. In a PL measurement at 10 K, the sample was excited by using different excitation powers of 2.4 μW , 10 μW , 20 μW , 60 μW , and 100 μW . The results are shown in Fig. 4. It can be seen that at low excitation powers, the spectrum shows a wider PL spectrum at the lower energy level and it can be fitted into two Gaussian curves. Further increasing the excitation power leads to the third peak, located at around 1.3 eV. It can be interpreted that the first two peaks at a lower excitation power arise from the QD with different sizes, and the third peak occurring

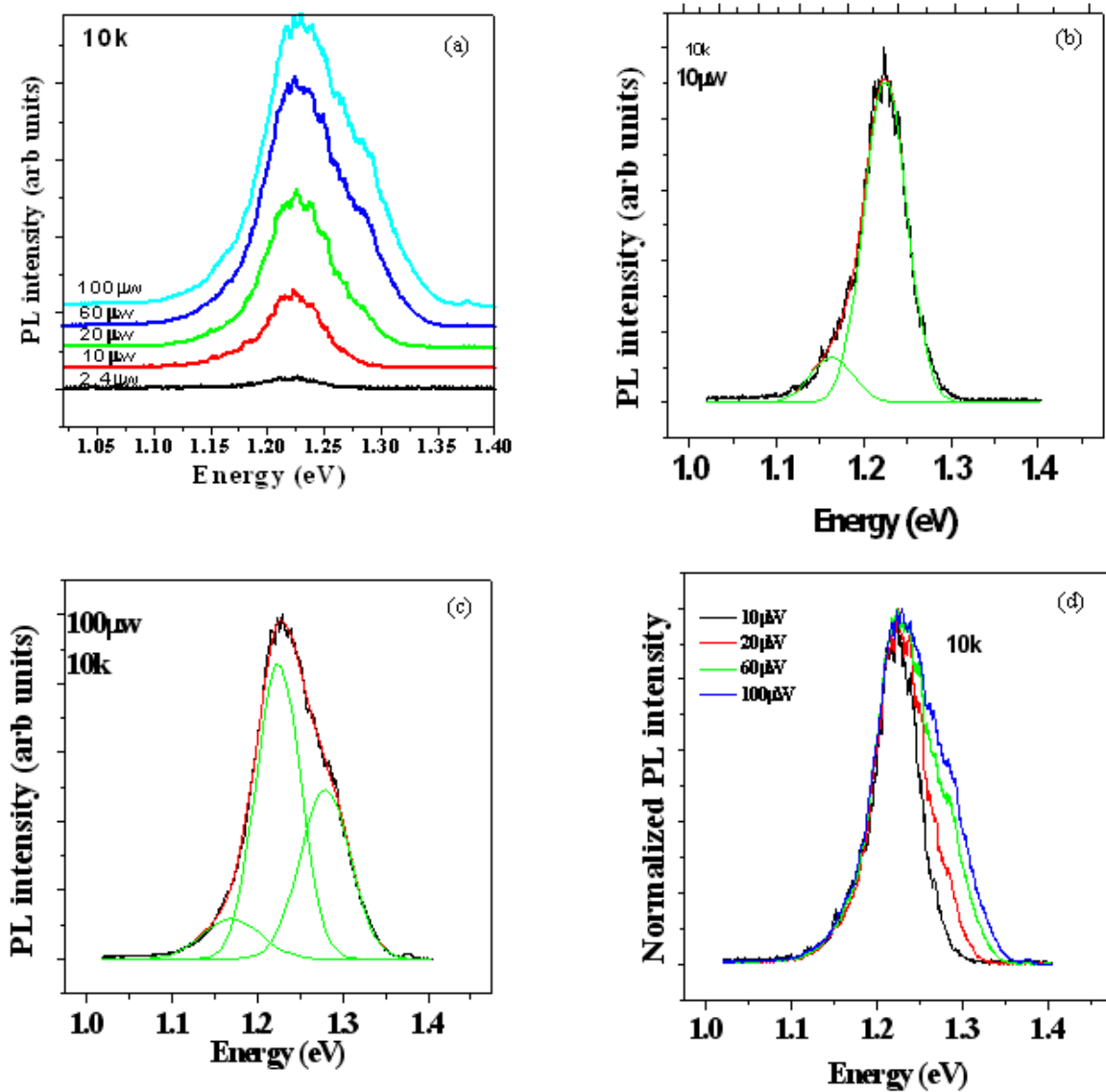
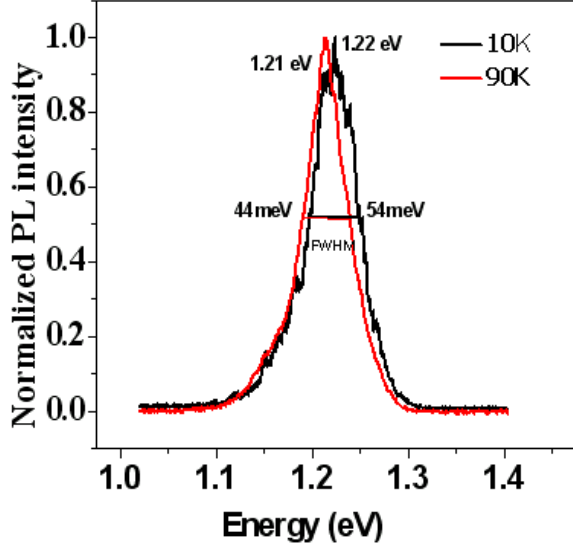


Fig.4: (a) excitation-power-dependent PL spectra, (b) a two-Gaussian-curve fit for the case of 10 W excitation power, (c) a three-Gaussian-curve fit for the case of 100 W excitation power, and (d) excitation-power-dependent normalized PL intensity.

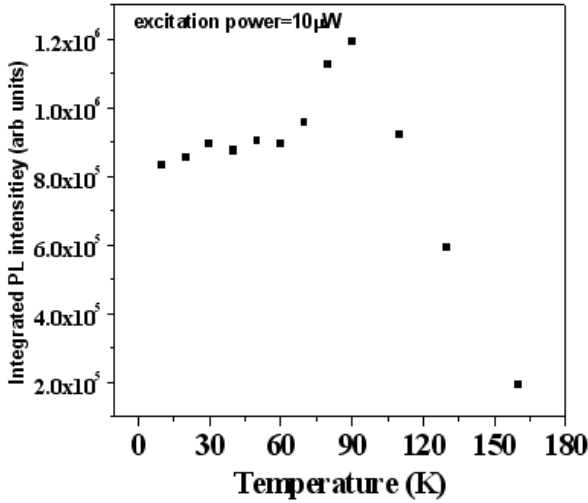
at a higher excitation power comes from the excited state of the QDs since both the first and the second peaks should be observable with the same intensity ratio [9], but in our case the intensity ratio of the second peak and the third peak is different when the excitation power was increased.

Fig. 5(a) shows temperature-dependent PL spectra of bi-QDs. It is seen that: First, the PL spectral shape and peak position are dependent on temperature. At 10 K, the PL peak was observed at 1.22 eV and when the temperature was increased to 90 K, the peak was shifted by 10 meV to the lower energy side. Second, the integrated PL intensity increases with the increasing temperature up to certain temperature and then decreases with further increasing the temperature.

In general, PL intensity decreases with increasing temperature due to an enhancement of non-radiative recombination process and a reduction of excitonic transition with temperature. One possible reason for an increase in the integrated PL intensity as well as a change of the peak position and the spectral line width is connected with the carrier dynamics. According to the AFM image, the quantum dots are close together and the distance between the dots is much smaller than size of the quantum dots. In this case the bi-QDs can be considered as coupled quantum dots. In case of a coupled quantum dot system, the wavefunctions of the carriers in the adjacent QDs overlap with each other. This overlapping of wavefunctions is expected to relieve the phonon relaxation bottle neck by increasing the number of states related



(a)

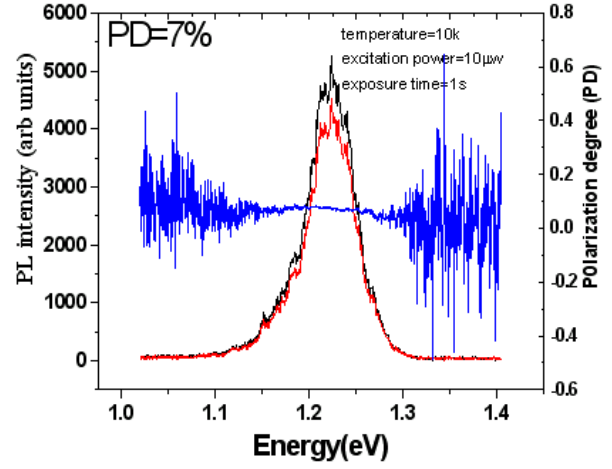


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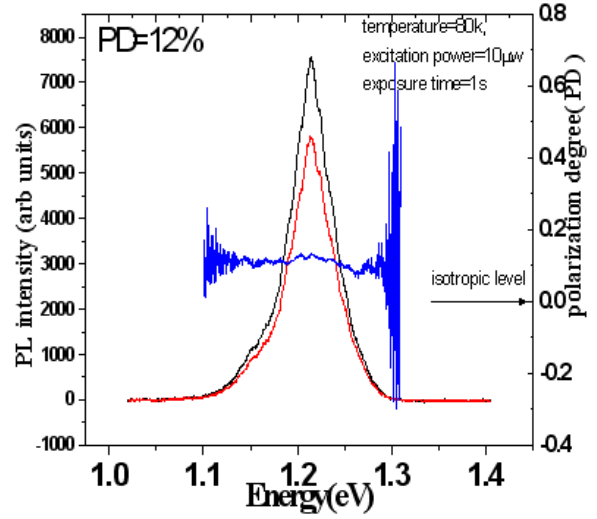
Fig. 5: (a) Temperature-dependent PL spectra at 10 K and 90K, and (b) integrated PL emission over the temperature range from 10 K to 160 K.

to carrier relaxation. When the temperature goes up, the coupling and relaxation effect will increase because of the increasing electron-phonon interaction. As a result, the photo-generated carriers transfer and relax into the energetically lower energy state, and recombine there. Consequently, the PL peak is shifted to a lower energy level. Increasing photo-generated carriers in the lower energy level causes a higher PL intensity at higher temperature. This process is also expected to cause a reduction in the FWHM of the PL spectrum. At 90 K, the integrated PL intensity is highest and the linewidth decreases to 44 meV compared with the 54 meV at 10 K. Further increasing the temperature, the linewidth increases again and the integrated PL intensity reduces. This is due to the electron-phonon scattering and thermal distribu-

tion [8-12].



(a)



(b)

Fig. 6: Temperature-dependent polarization-resolved PL spectra (a) at 10 K, and (b) at 90 K. Both spectra were obtained at the excitation power of 10 μ W and the measurement exposure time was 1 second.

The optical polarization anisotropy of the QD emission was also analyzed by observing the variation of PL signal intensity as the half-wave plate retarder orientation is rotated. The maxima and the minima were recorded when the analyzer axis was parallel and perpendicular to the orientation of the bi-QDs on the sample. Fig. 6 (a) and (b) show the polarization-resolved PL spectra of the bi-QD sample.

At 10 K, the linear polarization degree, defined as $PD = (I_{max} - I_{min}) / (I_{max} + I_{min})$, is only 7%; almost no polarization dependence was found for the PL emission from the bulk GaAs substrate, however. The anisotropy is nearly constant over the whole energy spectrum indicating a homogenous uniform shape, orientation and strain distribution [13]. Further increasing the temperature, the linear polar-

ization degree also increases, and at 50 K the linear polarization degree increases to a maximum value of 13%. Table 1 summarizes and compares some major different PL results obtained from measurements on bi-QDs at temperatures of 10 K and 90 K.

Table 1: Comparison on PL characteristics of bi-QDs measured at 10 K and 90 K.

Temperature	10 K	90 K
Peak Location	1.22 eV	1.21 eV
FWHM	54 meV	44 meV
Integrated PL Intensity	Lower	Higher
PD	7%	12%

Fig. 7 shows more information on the temperature-dependent linear polarization degree of bi-QDs. All polarization measurements were measured under non-resonant excitation in the barrier of GaAs and the time to measure the spectrum was 1 second.

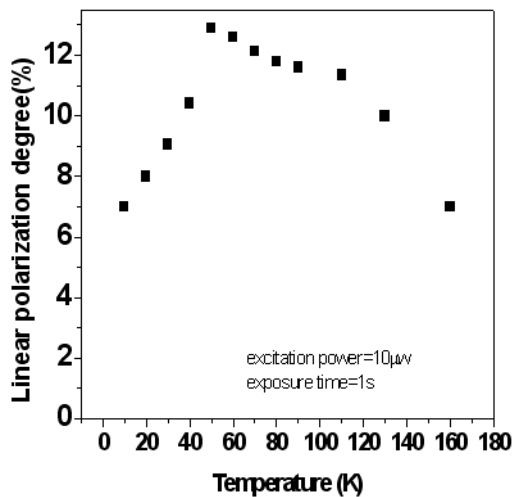


Fig. 7: Temperature-dependent linear polarization degree between 10 K and 160 K.

The temperature-dependent PL measurements show that the linear polarization degree (PD) changes with temperature. When the temperature increases, the PD increases at first and attains the highest degree around 50 K. Between 50 K and 90 K, the polarization degree is almost constant, and above 90 K the polarization degree starts to decrease sharply. Therefore, the polarization degree in this binary QD is probably related to the carrier dynamics in the QDs [14]. This kind of polarization behavior is probably caused by the thermal dislocation of carriers from 0-D quantum dots state to the 2-D quantum well state with the increasing temperature. At low temperatures, the excitons are localized in the QDs. When the temperature was increased to a certain point, the localized carriers obtain enough energy to travel through the two coupled QDs. The coupling strength

of the QDs becomes stronger and the QDs act like a 1-D quantum wire with an associated increase in polarization anisotropy. When the temperature is high enough, the carriers in the coupled QDs jumped into the nearest 2-D wetting layer and lost the directional dependence nature so that optical polarization anisotropy is reduced [15]. To reach the definite understanding of this mechanism, further investigation is needed.

5. CONCLUSION

Optical characteristics of bi-QDs were investigated. It was found that the integrated PL intensity, peak position, and the linear polarization degree depend on the excitation power and temperature. Such behaviors are related to the carrier dynamics.

6. ACKNOWLEDGEMENT

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quantum dot lasers (1999) and a vertical-cavity surface-emitting quantum-well laser (1999). He also achieved the first demonstration of the cavity-polariton in a semiconductor microcavity (1992). He is now regarded to be one of the most active scientific researchers in the field of semiconductor nanotechnologies. Particularly, he is intensively working on establishment core technologies of quantum dots and related nanostructures for optoelectronics applications. He is now the director of Nanoelectronics Collaborative Research Center, University of Tokyo, Japan.