Journal of the Korean Physical Society, Vol. 46, No. 6, June 2005, pp. 1396∼1400

Growth-Mode Transition Controlled by the Substrate Temperature in InAs/GaAs Quantum-Dot Ensembles

S. J. Lee, J. O. Kim, Y. G. Kim and S. K. Noh*
Quantum Dot Technology Laboratory, Korea Research Institute of Standards and Science, Daejeon 305-600

Y. H. Ryu and S. M. Choi
Department of Medical Research, Korea Institute of Oriental Medicine, Daejeon 305-811

J. W. Choe
Department of Electrical Engineering and Computer Science, Kyunghee University, Suwon 449-701

(Received 7 March 2005, in final form 6 April 2005)

We report a growth-mode transition observed in the optical and the structural characteristics of self-assembled InAs quantum-dot (QD) ensembles with different size distributions controlled by the substrate temperature. The photoluminescence spectra and the atomic force microscope images show that the QD ensembles change from a small-size mode to a large-size one via a bimodal phase appearing at a mode-transition temperature of 490 °C. On the bases of the variations of the emission energy and the apparent dimension, two kinds of growth mechanisms of adatom diffusion and island migration are suggested for the island ripening and the dynamic coalescence of QDs below and above the mode-transition temperature. Finally, we proposed a schematic model illustrating the size evolution of QDs.

PACS numbers: 68.65.Hb; 78.67.Hc; 81.07.Ta
Keywords: Quantum dot, Self-assembly, Photoluminescence, Atomic force microscope, Indium arsenide

I. INTRODUCTION

Particularly owing to strong confinement of the electron and the hole wave functions attributed to zero-dimensional (0D) features, self-assembled quantum dot (QD) structures have emerged as a novel system not only for explorations of exotic properties but also for applications of practical devices, such as QD-based memory devices [1], single photon emitters [2], laser diodes [3,4], and infrared photodetectors [5–7]. Although a number of experiments have been performed on self-assembled QD systems by changing the growth parameters and the constituent species/structures during the last decade, the controllability and the reproducibility still remain unsettled because of the complexity of the self-assembling growth kinetics [8–12]. Recently, some research groups reported doublet-like photoluminescence (PL) spectra and anomalous temperature dependences observed in bimodal and multimodal QD ensembles. Though the distinctive results appearing in the PL spectra and the atomic force microscope (AFM) images were nicely explained on basis of carrier transfer phenomenon arising in adjacent QDs, there was no clear mention of how the QD size mode evolved with the growth parameters [13–16]. One controversial issue remaining in QDs is the formation mechanisms related with growth-mode transitions, which must be solved in order to achieve reliable control of the sublevel energy, especially in association with the wavelength design of light emitters and detectors.

In order to understand the dependence of the growth kinetics on the growth temperature, in this study, we investigate the variations in the optical and the structural characteristics observed in a series of self-assembled InAs/GaAs QD structures with different size distributions controlled by the substrate temperature. Using a comparative analysis of the PL emission energy and the apparent AFM dimensions of the QD ensembles, we discuss two kinds of growth mechanisms for the distinctive domains arising below and above the mode-transition temperature (MTT) and for the bimodal phase appearing at the MTT. Finally, we propose a schematic model illustrating the dependence of the size evolution of QDs on the growth temperature.

II. EXPERIMENTAL PROCEDURE

For this study, a series of double-layered InAs-QD/GaAs structures grown at different temperatures were pre-

*E-mail: sknoh@kriss.re.kr

pared by using a molecular beam epitaxy (MBE) technique, and the spontaneous formation of InAs QDs was performed in the Stranski-Krastanow (S-K) mode. Five samples were grown under the same conditions, and basically, they had the same layer structure, except for the size distribution of the QDs, which varied with the substrate temperature. The buried QD layer was capped by 40 nm of GaAs for the PL measurement, and another QD layer was formed on a surface with no cap layer for the AFM analysis. The growth was initiated from a GaAs buffer layer with a thickness of 0.3 μm at a temperature of 550 °C, and the QD formations were conducted on the buffer layer at lower temperatures in the range of 470 – 505 °C. The equivalent thickness and the growth rate of the InAs QDs were 2.3 monolayers (MLs) and 0.08 ML/s, respectively, and the V/III beam-equivalent pressure (BEP) was fixed at approximately 38. The dot formation was *in-situ* confirmed by observing the 2D-3D transition in the reflection high-energy electron diffraction (RHEED) pattern, and the dot profiles were analyzed by using AFM images.

PL measurements were performed by using a typical visible-to-near-infrared monochromator system with an Ar-ion laser (λ = 514.5 nm) and a closed-cycle He refrigerator (10 K). Excitation-power dependent PL spectra were taken in the range of nominal output power of 0.1 – 100 mW at 20 K, and the luminescence signals were detected and recovered by using a thermoelectric-cooled InGaAs photodiode and a lock-in amplifier. The QD dimensions were histographically displayed by using the data analyzed from a hundred QD images randomly sampled in each AFM profile. The epitaxial growth and the basic properties of similar QD heterostructures have been reported elsewhere [7,13].

III. RESULTS AND DISCUSSION

Fig. 1 shows two series of representative PL spectra (20 K) taken at nominal excitation powers of (a) 1 mW (low power) and (b) 100 mW (high power) for five samples grown at different temperatures. The PL spectra clearly show one or two dominant peaks associated with InAs QD emissions around 1.1 eV and 1.2 eV. While the two samples grown at low temperatures (470 °C, 480 °C) show a single peak with no power dependence, the other two samples grown at high temperatures (500 °C, 505 °C) have doublet-like peaks with strong power dependences, indicating that the high-energy peak is an excited state. These suggest that all the QD ensembles of the four samples have monomodal size distributions. On the other hand, only the PL spectrum for a QD sample fabricated at an inbetween temperature of 490 °C is similar to those of the high-temperature samples, except for a weak power dependence that implies a bimodal size distribution [13]. Taking account of the power-dependent behaviors of the PL spectra, we can say that, as the growth temperature is increased, the QD ensemble gradually changes from a small QD mode with high emission energy (1.15 $\sim$ 1.2 eV) to a large QD mode with low emission energy ($\sim$ 1.1 eV), passing through a bimodal phase, in which small QDs and large QDs coexist, near a mode-transition temperature (MTT) of $\sim$ 490 °C. Hereinafter, we refer to the QDs responsible for the high-energy and the low-energy PL peaks as *small QDs (SQDs)* and *large QDs (LQDs)*, respectively. Consequently, the doublet-like peak shown in high-temperature samples originates from a ground state (GS) and an excited state (XS) of a LQD whereas the single peak appearing in the low-temperature samples is attributed to a GS(SQD). In addition, the bimodal peak formed at MTT consists of a GS(LQD) and a mixed state overlapped by GS(SQD) and XS(LQD), as denoted in Fig. 1. The reason no XS(SQD) is observed in those PL spectra is that the sublevel is high enough for it not to be confined in the InAs-QD well.

It seems a little strange for the PL intensity of GS(LQD) in the bimodal sample to be still much stronger than that of GS(SQD) even for high excitation power. However, this behavior can be understood by us-

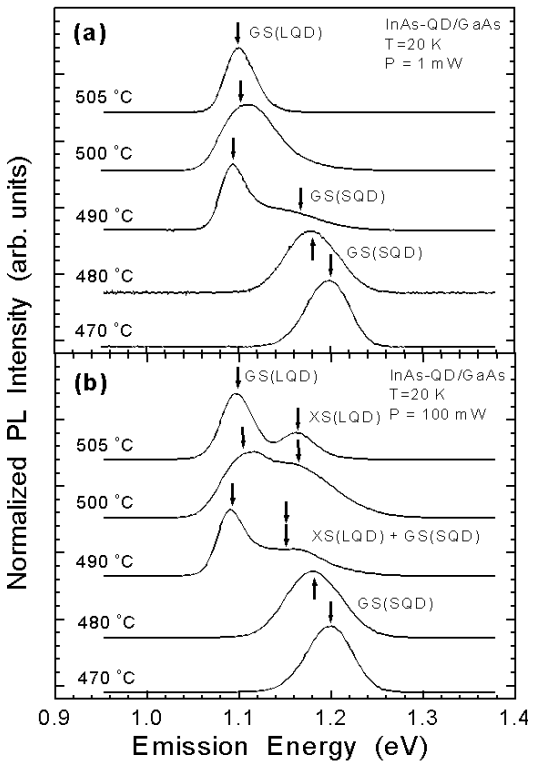

Fig. 1. Growth-temperature dependent PL spectra (20 K) taken at excitation powers of (a) 1 mW (low power) and (b) 100 mW (high power) for five samples grown at different temperatures. The PL spectra clearly show one or two dominant peaks associated with InAs QD emissions around 1.1 eV and 1.2 eV. The three samples grown at temperatures above 490 °C show excitation-power dependences whereas the others show no dependence.

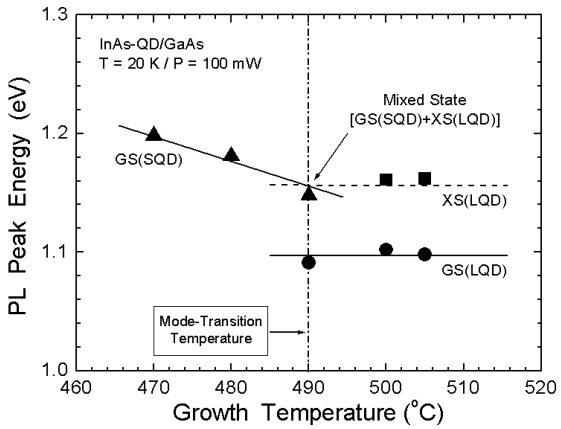

Fig. 2. PL peak energy as a function of the growth temperature. The GS(SQD) energy gradually decreases up to MTT, but the energies of GS(LQD) and XS(LQD) are almost constant above MTT.

ing the carrier redistribution mechanism [14]. In bimodal QD systems, it may be thermodynamically possible for carrier repopulation between adjacent QDs through the wetting layer as a carrier transfer channel even at an appropriately low temperature. In the case of the bimodal sample of Fig. 1, the electrons activated from GS(SQD) with higher energy can transfer to GS(LQD) with lower energy; thus, the PL intensity of GS(LQD) becomes much stronger than that of GS(SQD). It is worth noting that for subsequent discussions of the AFM profiles, the low-energy QD usually has a stronger PL intensity than the high-energy QD, even in the case when the number of SQDs is comparable to that of LQDs.

A graph showing the PL peak energies as a function of the growth temperature is presented in Fig. 2. All the energy values shown in the graph were determined by using two-peak Gaussian-curve fits. As the temperature was increased, the GS(SQD) energy gradually decreased up to MTT, but the energies of GS(LQD) and XS(LQD) are almost constant at (1.097 ± 0.005) eV and (1.167 ± 0.007) eV above MTT. These show that the QD size gradually develops with increasing growth temperature and that the effective dimension associated with PL emission tends to become saturated via a bimodal QD phase formed at a specific temperature of MTT. In order to support the evolutionary dependence of the PL peak energies on the growth temperature, we tried a quantitative analysis by AFM profiling.

Fig. 3(a) shows AFM images of the surfaces of the same samples used in the PL measurements, and Fig. 3(b) shows QD size distributions for the lateral diameter (left) and the vertical height (right). Each histogram was plotted by using the values obtained from cross-sectional profiles of a hundred QDs randomly selected in the corresponding AFM figure. A distinguishing feature was found in the bimodal sample. Many twins coupled by adjacent QDs were observed, as shown in the inset of the middle AFM picture of Fig. 3(a). In this case, the

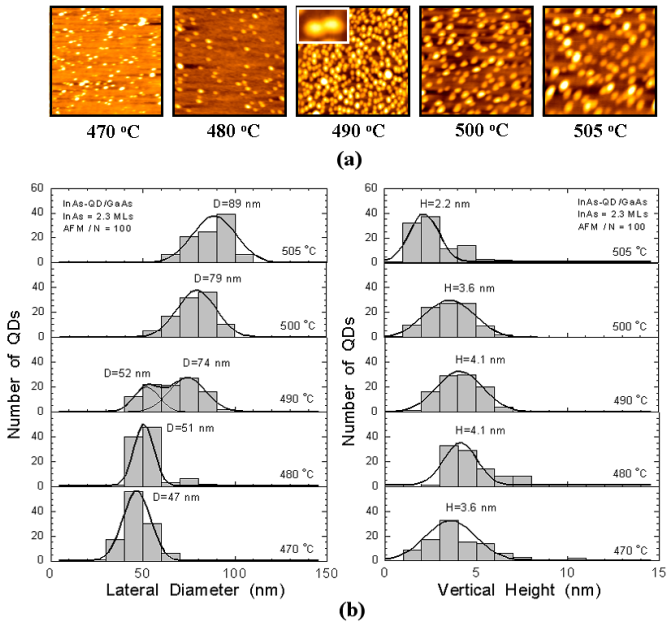

Fig. 3. (a) AFM images for the same samples used for the PL measurements. (b) QD size distributions for the lateral diameter (left) and the vertical height (right). Each histogram was plotted by using the values obtained from cross-sectional profiles of a hundred QDs randomly selected in the corresponding AFM figure.

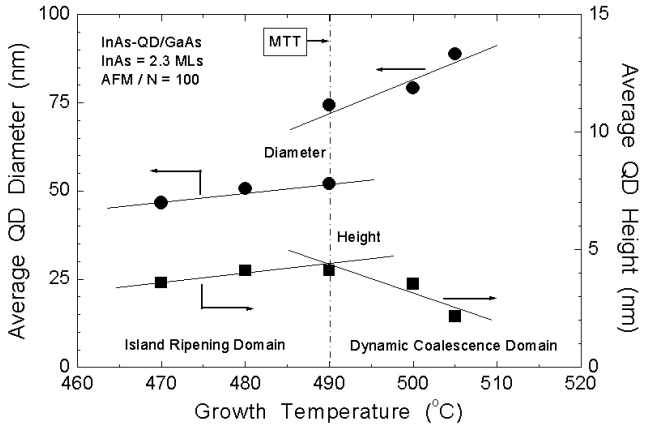

Fig. 4. Average diameters and heights determined by using the Gaussian fits of the histograms as a function of the growth temperature. Both the diameters and the heights simultaneously become larger up to MTT, but the heights get smaller where the diameters continue to get larger.

longitudinal length of the twins was taken as diameter of a QD. That the Gaussian-fit diameter curve of the bimodal sample has two maxima (D = 52 nm and D = 74 nm) is an expression of the coexistence of isolated single and coupled twin QDs. In addition, the sudden increase in the QD density appearing in the bimodal sample is a result of the formation of twin QDs counted as two single QDs. This behavior can not be simply interpreted by using a specific mechanism, but only by using a transient

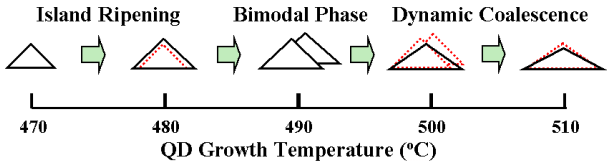

Fig. 5. Schematic model, based on the PL and the AFM results, showing the size evolution of QDs.

phase formed between two different growth modes, which will be discussed in the final paragraph of this paper.

The average diameters and heights determined from the Gaussian-curve fits superposed on the histograms of Fig. 3(b) are plotted in Fig. 4 as functions of the growth temperature. Both the diameters and the heights simultaneously become larger up to MTT, but the height is reduced a little above MTT, and the diameter is increased. The size variation of the SQD samples below MTT is fairly consistent with that discussed in the PL spectra. The size enhancement of QDs gives rise to a lowering of QD sublevel that results in a decrease of the PL energy, as expected in normal quantum well or dot structures. However, the dimensional variations of the LQD samples somewhat conflict with those described for the PL spectra. In contrast to the constant PL energy suggesting no size change in LQDs, AFM images show rapid increases in the diameters and slight decreases in the heights. The inconsistency between the apparent dimension and the emission energy can be interpreted by simultaneously considering the variations of diameter and height. It has been generally accepted that the energy levels of QDs more strongly depend on height than on diameter. Thus, the size effect of diameter enhancement can be balanced by that of height reduction so as to keep a constant sublevel energy, similarly to the results plotted in Fig. 2.

The structural evolution demonstrated in the AFM profiles suggest that two kinds of growth domains ruled by distinctive mechanisms exist below and above MTT. One is the island ripening domain available for low-temperature growth, and the other the dynamic coalescence domain, which is expected for high-temperature growth. In case of sufficiently low growth rate as in this study, the size distribution of QD ensembles is known to strongly depend on the growth temperature and to mostly develop through two kinds of processes, thermal diffusion of adatoms and dynamic migration of islands [17, 18]. Since the Ostwald ripening process through adatom diffusion may be dominant in the low-temperature region, one can expect simultaneous increases in the diameter and the height, which give rise to a decrease in the PL emission energy of QDs. In the high-temperature region, on the other hand, the dynamic coalescence process may become important due to island migration, which results in an increase in the diameter and a decrease in the height. Because the self-assembled QDs are thermodynamically unstable rather than in a minimum configuration of the Gibbs free energy of strained islands, it is very difficult to explain the transient behavior of the twin QDs generated at the MTT. At present, we understand that the bimodal phase in which single and twin QDs coexist may be attributed to a competition between adatom diffusion and island migration happening at a specific temperature. Thus, we can conclude that the SQD growth mode changes to the LQD one with increasing growth temperature, passing through a transient bimodal phase of single and twin QDs which is formed at a specific temperature. Fig. 5 is a schematic model showing the size evolution of QDs discussed above.

IV. SUMMARY AND CONCLUSIONS

We presented PL spectra and AFM images taken from five self-assembled InAs QD ensembles with different size distributions controlled by the substrate temperature. The PL spectra showed a bimodal spectral feature at an MTT of 490 °C, and the QD energy slowly decreased and became saturated at a constant value with enhancing growth temperature. The AFM profiles revealed that both the diameter and the height simultaneously became larger up to MTT above which the diameter increased but the height decreased. The optical and the structural characteristics showed that the QD ensembles gradually evolved from an SQD growth mode to a LQD one via a bimodal phase of single and twin QDs formed at a specific temperature. On the bases of the variations of the emission energy and the apparent dimension, the diameter enhancement effect could be balanced by the height reduction effect so as to keep a constant sublevel energy, and the QD energy more strongly depended on height than on diameter. Two kinds of growth mechanisms, adatom diffusion and island migration, had been suggested for the island ripening and the dynamic coalescence of QDs arising below and above MTT, respectively. Finally, we proposed a schematic model illustrating the size evolution of QDs based on the growth mechanisms discussed in this study.

ACKNOWLEDGMENTS

This work was carried out at the National Research Laboratory on Quantum-Dot Technology at the Korea Research Institute of Standards and Science nominated by the Ministry of Science and Technology (Contract No. M1-0104-00-0127). One of authors (SKN) acknowledges the partial support provided by the Korea Science and Engineering Foundation through the Quantum-functional Semiconductor Research Center at Dongguk University.

REFERENCES

[1] P. M. Petroff, A. Lorke and A. Imanoglu, Phys. Today **54**, 46 (2001).

[2] Z. Yuan, B. E. Kardynal, R. M. Stevenson, A. J. Shields, C. J. Lobo, K. Cooper, N. S. Beattie, D. A. Ritchie and M. Pepper, Science **295**, 102 (2002).

[3] M. Kuntz, G. Fiol, M. Laemmlin, D. Bimberg, M. G. Thompson, K. T. Tan, C. Marinelli, R. V. Penty, I. H. White, V. M. Ustinov, A. E. Zhukov, Yu. M. Shernyakov and A. R. Kovsh, Appl. Phys. Lett. **85**, 843 (2004).

[4] I. R. Sellers, H. Y. Liu, K. M. Groom, D. T. Childs, D. Robbins, T. J. Badcock, M. Hopkinson, D. J. Mowbray and M. S. Skolnick, Electron. Lett. **40**, 1412 (2004).

[5] J. Jiang, K. Mi, S. Tsao, W. Zhang, H. Lim, T. O'Sullivan, T. Sills, M. Razeghi, G. J. Brown and M. Z. Tidrow, Appl. Phys. Lett. **84**, 2232 (2004).

[6] E. -T. Kim, A. Madhukar, Z. Ye and C. Campbell, Appl. Phys. Lett. **84**, 3277 (2004).

[7] S. K. Kang, S. J. Lee, J. I. Lee, M. D. Kim, S. K. Noh, Y. H. Kang, U. H. Lee, S. C. Hong, H. S. Kim and C. G. Park, J. Korean Phys. Soc. **42**, 418 (2003).

[8] B. A. Joyce and T. B. Joyce, J. Cryst. Growth **264**, 605 (2004).

[9] J. L. Movilla, J. I. Climente and J. Planelles, J. Appl. Phys. **94**, 4515 (2003).

[10] V. G. Dubrovskii, G. E. Cirlin, Yu. G. Musikhin, Yu. B. Samsonenko, A. A. Tonkikh, N. K. Polyakov, V. A. Egorov, A. F. Tsatsul'nikov, N. A. Krizhanovskaya, V. M. Ustinov and P. Werner, J. Cryst. Growth **267**, 47 (2004).

[11] R. Songmuang, S. Kiravittaya and O. G. Schmidt, J. Cryst. Growth **249**, 416 (2003).

[12] H. Rho, J. D. Song, Y. J. Park, W. J. Cho and J. I. Lee, J. Korean Phys. Soc. **45**, 149 (2004).

[13] S. J. Lee, S. K. Noh, J. W. Choe and E. K. Kim, J. Cryst. Growth **267**, 405 (2004).

[14] L. Brusaferri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri and S. Franchi, Appl. Phys. Lett. **69**, 3354 (1996).

[15] H. Lee, R. Lowe-Webb, T. J. Johnson, W. Yang and P. C. Sercel, Appl. Phys. Lett. **73**, 3556 (1998).

[16] H. Kissel, U. Mueller, C. Walther, W. T. Masselink, Yu. I. Mazur, G. G. Tarasov and M. P. Lisitsa, Phys. Rev. B**62**, 7213 (2000).

[17] Y. Nakata, K. Mukai, M. Sugawara, K. Ohtsubo, H. Ishikawa and N. Yokoyama, J. Cryst. Growth **208**, 934 (2000).

[18] P. B. Joyce, T. J. Krzyzewski, G. R. Bell, T. S. Jones, S. Malik, D. Childs and R. Murray, Phys. Rev. B **62**, 10891 (2000).