Temperature dependence of photoluminescence characteristics of excitons in stacked quantum dots and quantum dot chains

Kojima, Osamu
Nakatani, Hiroaki
Kita, Takashi
Wada, Osamu
Akahane, Kouichi

(Citation)
Journal of Applied Physics,107(7):073506-073506

(Issue Date)
2010-04-01

(Resource Type)
journal article

(Version)
Version of Record

(URL)
https://hdl.handle.net/20.500.14094/90001171

Temperature dependence of photoluminescence characteristics of excitons in stacked quantum dots and quantum dot chains

Osamu Kojima,[1,a)] Hiroaki Nakatani,[1] Takashi Kita,[1] Osamu Wada,[1] and Kouichi Akahane[2]
[1]*Department of Electrical and Electronic Engineering, Graduate School of Engineering, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan*
[2]*National Institute of Information and Communications Technology, 4-2-1 Nukui-kitamachi, Koganei, Tokyo 184-8795, Japan*

(Received 15 September 2009; accepted 18 February 2010; published online 7 April 2010)

We have investigated the effects of temperature on the photoluminescence (PL) characteristics of excitons in ordinary stacked quantum dots (QDs) and QD chains in which QDs are interconnected along the growth direction. While the temperature dependence of the PL intensity of both samples is similar, that of the PL decay time is different. In addition, the PL decay times of both samples monitored at 150 K clearly depend on the detection energy. This result is attributed to lateral QD coupling. From these results, in ordinary stacked QDs, the exciton transfer owing to the lateral coupling is the only cause of the increase in the PL decay time. On the other hand, in QD chains, the interconnection along the chain direction as well as the lateral coupling is considered to cause the change in the PL characteristics and induce the extremely long exciton lifetime. © *2010 American Institute of Physics*. [doi:10.1063/1.3366711]

I. INTRODUCTION

Quantum dots (QDs) offer the possibility of realizing various interesting devices such as QD lasers,[1] ultrafast optical switches,[2,3] and solar cells.[4,5] To realize of such QD devices, it is necessary to design the optical properties by controlling the oscillator strength of excitons and by fabricating high-quality and high-density QDs. From this viewpoint, we clarified that the photoluminescence (PL) intensity, PL energy, and exciton lifetime in multiple stacked QDs (SQDs) fabricated by using the strain compensation technique[6] can be controlled by changing the QD separations along the growth direction.[7,8] In the SQD structures, the overlap of electron envelope functions between QDs along the growth direction increases as the spacer layer thickness decreases, leading to the interconnection of QDs. This results in a considerable change in the PL characteristics due to the change in the exciton oscillator strength. Then, we can regard such QDs interconnected with each other via electron envelope functions along the growth direction as QD chains (QDCs), and distinguish them from ordinary SQD ensembles that do not involve any interaction along the growth direction.

The reduced oscillator strength of excitons in a QDC structure may not be suitable for light-emission devices such as LEDs, laser devices, and so on without a further increase in the stacking layer. However, some devices such as quantum information devices and solar cells require a long exciton lifetime. In particular, in QD solar cells, a long exciton lifetime improves the efficiency of electron output. Thus, the controllable long exciton lifetime found in our previous work is considered to be a noteworthy property of QDCs for realizing novel devices. Here, we focus on a carrier dynamics depending on the temperature in order to clarify the effects of the interconnection in vertically aligned QDCs. In this study, we have investigated the temperature dependence of the PL characteristics of SQDs and QDCs that were fabricated by using the strain compensation technique. While the temperature dependence of the PL intensity of both samples is similar, that of the PL decay time is different. In addition, the detection-energy dependence of the PL decay time at 150 K in both samples indicates the occurrence of exciton transfer from smaller QDs to larger ones in each layer. We discuss the temperature dependence of the PL characteristics of SQDs and QDCs on the basis of the interaction between the QDs in the lateral and vertical directions.

II. EXPERIMENT

We used two samples in this study. For each sample, InAs self-assembled QDs with 30 layers were grown on an InP(311)B substrate by molecular beam epitaxy using a strain compensation technique.[8] We deposited 4 ML InAs QDs on a 150 nm thick $In_{0.52}Al_{0.48}As$ buffer layer. The samples have $In_{0.5}Ga_{0.1}Al_{0.4}As$ spacer layers with thicknesses of 20 and 40 nm. The QD density in each sample is $2.5\times10^{12}/cm^2$. According to our previous report,[8] a sample with a spacer layer thickness of 40 nm corresponds to an SQD, and that with a thickness of 20 nm corresponds to a QDC, in which the QDs are interconnected along the growth direction. The spacer layer compensates for the stress caused by the lattice mismatch between the QD layer and the buffer layer. We measured the PL characteristics at temperatures between 3.4 and 180 K. Conventional PL was measured by using the 488 nm line of a CW Ar^+ laser. The excitation density was kept at 100 W/cm^2. The emitted light was dispersed by a 32 cm single monochromator with a resolution of 1.0 nm and was detected by using a liquid-nitrogen-cooled InGaAs-photodiode array. The PL decay profiles were measured by using a time-correlated single-photon counting

a)Electronic mail: kojima@phoenix.kobe-u.ac.jp.

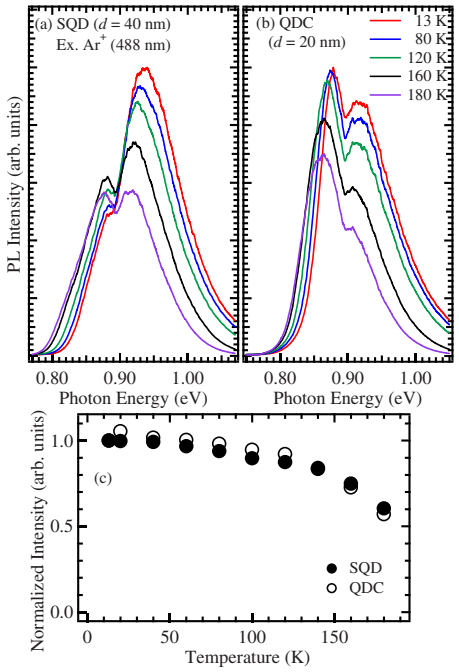

FIG. 1. (Color online) Temperature dependence of the PL spectra in (a) SQD and (b) QDC samples. The dip at around 0.9 eV originates from the hydroxy group in the optical fiber. (c) The normalized integrated PL intensities in both the samples were plotted as a function of temperature.

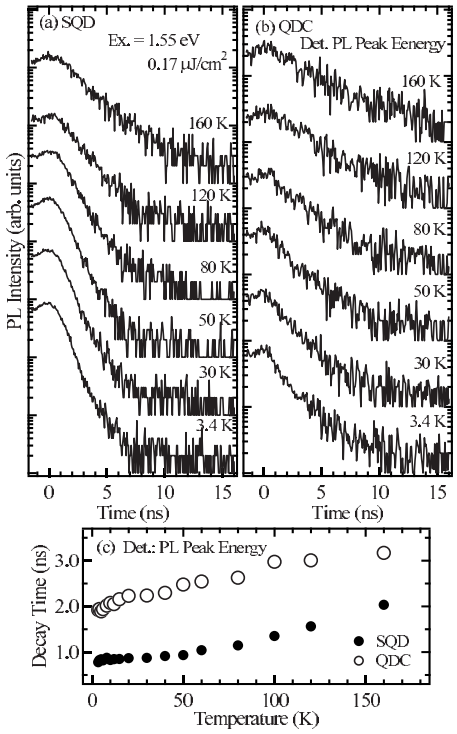

FIG. 2. Temperature dependence of the PL decay profiles in (a) SQD and (b) QDC samples. Each profile was recorded at the PL peak energy. (c) The PL decay times in both the samples are plotted as a function of temperature.

method with a time resolution of 0.8 ns. The excitation source was a mode-locked Ti:sapphire pulse laser delivering 110 fs pulses with a repetition rate of 4 MHz. We used a pulse picker to reduce the laser repetition rate from 80 to 4 MHz. The excitation photon energy was 1.550 eV and the excitation density was kept at 0.17 $\mu J/cm^2$. The PL was dispersed by using a 27 cm single monochromator with a resolution of 1.0 nm and was detected using a time-to-amplitude converter system with a liquid-nitrogen-cooled InP/InGaAsP photomultiplier.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the PL spectra for both the samples. All spectra were normalized by the maximum intensity of the spectra at 13 K in each sample. The dip at around 0.9 eV is due to absorption by the hydroxy group in the optical fiber. The asymmetrical profile of the PL band in the QDC sample is considered to be caused by the QD interconnection.[8] According to the results of the detection-energy dependence of the PL decay time, the peak at 0.95 eV arises not from the higher exciton states but from the SQD component. As shown in Fig. 1(c), the temperature dependence of the integrated PL intensity in both the samples, where the intensities were normalized by that at 13 K, exhibits similar profiles. This result indicates that the nonradiative recombination process induced by an increase in the temperature is similar in both the samples.

Moreover, the PL intensity in the QDC sample increases slightly in the low temperature region. Assuming the strain distribution calculated by Grundmann in and around a pyramidal InAs QD,[9] there exist lateral potentials for electrons and holes in the vicinity of a dot. The potential for holes increases close to the dot and gives rise to a barrier for the capture of holes from the wetting layer (WL) in the QD. On the other hand, the potential for electrons drops monotonically due to the weak influence. Therefore, the excited carriers in the WL lead to the transfer of an impaired hole into the QDs, which reduces the PL quantum efficiency at low temperature. Because the hole potential exceeds that by thermal energy at 20 K, the PL intensity increases. In the case of the SQD sample, the thickness of the WL is greater than that in the QDC sample. We could not observe the increase in the PL intensity in the SQD sample due to this difference.

In order to clarify the difference between the temperature dependences in both samples, we measured the PL decay profiles at various temperatures, as shown in Fig. 2. Here, we defined a time of zero nanosecond to coincide with the PL peak. All the profiles were recorded at the PL peak energy. The PL decay times τ of both samples increase with the temperature. In Fig. 2(c), the τ values of both samples are plotted as a function of temperature. The increase in τ indicates that the growth by the strain compensation technique can suppress the generation of nonradiative centers with QD stacking. However, the temperature dependence of the PL decay time in the QDC sample is different from that in the SQD sample.

The increase in τ with the temperature is generally explained by the thermal dissociation of excitons into the electron-hole pairs, in which the excitons escape from the QDs into the spacer layer and/or the upper subband levels of electron and holes via thermionic emission.[10–13] We considered that the temperature dependence of the PL decay time in the SQD sample can be explained by this model. However, the temperature dependence of the PL decay time in the

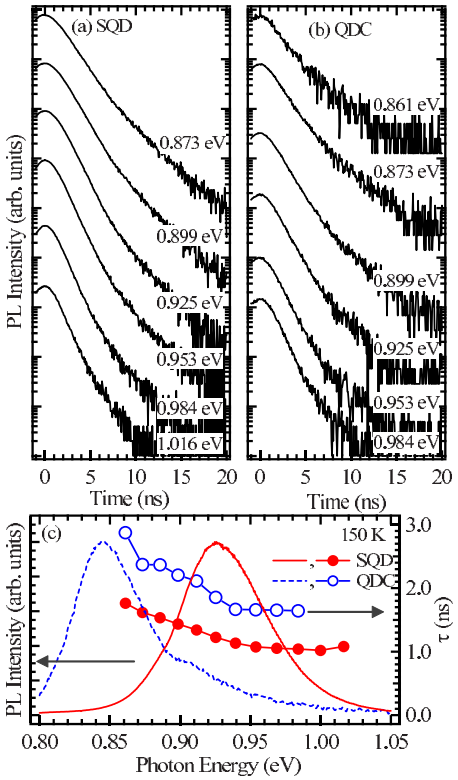

FIG. 3. (Color online) Detection-energy dependence of the PL decay profiles in (a) SQD and (b) QDC samples. (c) Detection-energy dependence of τ and PL spectra at 150 K.

QDC sample should include another factor, namely, the interconnection effect of the electron envelope functions.

To clarify the effect of the thermal dissociation, we examined the detection-energy dependence of the PL decay profiles at 150 K. The PL decay profile at each detection energy is shown in Figs. 3(a) and 3(b). Clearly, the τ values in both the samples depend on the detection energy. Figure 3(c) summarizes τ plotted as a function of the detection energy. The τ values were evaluated by fitting single exponential function form for the signals after five nanosecond to eliminate the distortion of signal around zero nanosecond. The PL spectra at 150 K are also shown for reference. As mentioned in our previous paper, τ at a low temperature of 3.4 K is constant in each sample.[8] The results obtained at 150 K differ considerably from those at a low temperature. This significant detection-energy dependence can be attributed to lateral coupling between QDs.[14,15]

The lateral coupling is caused by exciton transfer from smaller QDs to larger ones. The base structure of our QDs is an ellipse with a typical size of 39×51 nm^2, and the average distance between QDs is approximately 50 nm.[6] Thus, the bases of the QDs are very close to each other. Because the lateral distance between QDs is shorter than the separation in the growth direction, the exciton transfer is thermally triggered in the lateral direction. This exciton transfer could be observed as the PL rise time in the decay profile. Unfortunately, we could not observe the temporal evolution of the PL intensity caused by such exciton transfer because of the limitation of the time resolution of the photomultiplier. In the SQD sample, therefore, the interaction between the QDs is considered to be restricted along the in-plane direction, so that the PL characteristics can be explained by the thermal dissociation model. In other words, thermally delocalized excitons in the SQDs can transfer to the larger QDs within the same layer, and the excitons relax thereafter. On the other hand, in the QDC sample, the electron envelope functions of excitons localized in the QDs are elongated along the growth direction due to the interconnection after the transfer, inducing an extremely long exciton lifetime. Moreover, the elongation of the envelope function leads to the difference in the temperature dependence of τ. Our findings indicate that the QD interconnection in the growth direction occurs even at 150 K and affects the PL characteristics of the QDC.

Finally, in Figs. 2(a) and 2(b), the decay profiles exhibit intense signals in the negative time region. These components may indicate possible intraband transition and/or exciton transfer processes. However, we consider that the time resolution of the detector includes an ambiguity for the time of zero. Therefore, to clarify the origins of the signal in the negative time region, further studies using a system with superior time resolution are required.

IV. CONCLUSION

We have investigated the effects of temperature on the PL characteristics of excitons in SQDs and QDCs fabricated using the strain compensation technique. We found that the decrease in the PL intensity in the QDC sample is similar to that in the SQD sample. To clarify the effect of the interconnection in the QDC sample, we examined the temperature dependence of the PL decay time. The PL decay profiles, which show the increase in τ with temperature, indicated the suppression of nonradiative recombination paths caused during the QD and spacer layer growth processes. The increase in τ arises from the thermal delocalization. However, the temperature dependence of τ in the two samples is different. In order to reveal the discrepancy in the temperature dependence of the PL characteristics, we examined the detection-energy dependence of τ. The τ values of both samples clearly depend on the detection energy; this indicates the lateral coupling between the QDs. As the temperature increases, the excitons transfer from smaller QDs to larger ones. This affects the exciton relaxation process. However, in the QDC sample, the vertical interaction in addition to the lateral interaction strongly affects the excitonic process, and therefore, the temperature dependence of τ differs from that in the SQD sample. These findings suggest that the interconnection of QDs along the growth direction via the overlapping of electron envelope functions occurs at high temperatures. These may aid the development of some functional devices by using QDCs. In particular, they will be advantageous for QD solar cells based on the so-called QD superlattice.

ACKNOWLEDGMENTS

This work was partially supported by Grants-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT) of Japan, for Scientific Research on Innovative Areas "Optical Science of Dynamically Correlated Electrons" from MEXT of Japan, the

Inc. Administrative Agency New Energy and Industrial Technology Development Organization, and Ministry of Economy, Trade and Industry, Japan.

[1]D. L. Huffaker, G. Park, Z. Zou, O. B. Shchekin, and D. G. Deppe, Appl. Phys. Lett. **73**, 2564 (1998).
[2]R. Prasanth, J. E. M. Haverkort, A. Deepthy, E. W. Bogaart, J. J. G. M. van der Tol, E. A. Patent, G. Zhao, Q. Gong, P. J. van Veldhoven, R. Nötzel, and J. H. Wolter, Appl. Phys. Lett. **84**, 4059 (2004).
[3]E. W. Bogaart, R. Nötzel, Q. Gong, J. E. M. Haverkort, and J. H. Wolter, Appl. Phys. Lett. **86**, 173109 (2005).
[4]A. Martí, E. Antolín, C. R. Stanley, C. D. Farmer, N. López, P. Díaz, E. Cánovas, P. G. Linares, and A. Luque, Phys. Rev. Lett. **97**, 247701 (2006).
[5]R. Oshima, A. Takata, and Y. Okada, Appl. Phys. Lett. **93**, 083111 (2008).
[6]K. Akahane, N. Ohtani, Y. Okada, and M. Kawabe, J. Cryst. Growth **245**, 31 (2002).
[7]H. Nakatani, T. Kita, O. Kojima, O. Wada, K. Akahane, and M. Tsuchiya, J. Lumin. **128**, 975 (2008).
[8]O. Kojima, H. Nakatani, T. Kita, O. Wada, K. Akahane, and M. Tsuchiya, J. Appl. Phys. **103**, 113504 (2008).
[9]M. Grundmann, O. Stier, and D. Bimberg, Phys. Rev. B **52**, 11969 (1995).
[10]G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz, and P. M. Petroff, Appl. Phys. Lett. **64**, 2815 (1994).
[11]H. Yu, S. Lycett, C. Roberts, and R. Murray, Appl. Phys. Lett. **69**, 4087 (1996).
[12]A. Fiore, P. Borri, W. Langbein, J. M. Hvam, U. Oesterie, R. Houdré, R. P. Stanley, and M. Ilegems, Appl. Phys. Lett. **76**, 3430 (2000).
[13]R. Hostein, A. Michon, G. Beaudoin, N. Gogneau, G. Patriache, J.-Y. Marzin, I. Robert-Phillip, I. Sagnes, and A. Beveratos, Appl. Phys. Lett. **93**, 073106 (2008).
[14]A. Tackeuchi, Y. Nakata, S. Muto, Y. Sugiyama, T. Usuki, Y. Nishikawa, N. Yokoyama, and O. Wada, Jpn. J. Appl. Phys., Part 1 **34**, L1439 (1995).
[15]Y. I. Mazur, B. L. Liang, Zh. M. Wang, G. G. Tarasov, D. Gunzun, G. J. Salamo, T. D. Mishima, and M. B. Johnson, J. Appl. Phys. **100**, 054313 (2006).