Strong Photoluminescence Emission from an Excited-Subband Exciton State in a GaAs/Al$_x$Ga$_{1-x}$As Triple Quantum Well with Different Well Thicknesses

A. Satake[1*], N. Shiraishi[1], N. Takata[1], K. Fujiwara[1], L. Schrottke[2], and H. T. Grahn[2]
[1]Department of Electrical Engineering, Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan
[2]Paul Drude Institute for Solid-State Electronics, Hausvogteiplatz 5-7, 10117 Berlin, Germany
[*]Corresponding author: Phone: +81-93-884-3240 Fax: +81-93-884-0879 Email: satake@ele.kyutech.ac.jp

Abstract: We have studied the photoluminescence (PL) properties of a GaAs/Al$_x$Ga$_{1-x}$As quantum well (QW) system consisting of three QW's with different thicknesses, in which the widest one (7.8 nm) has an excited-subband exciton state located slightly below the barrier band edge. Using above-barrier excitation with a power density of about 10 W/cm^2 at 13 K, a strong PL emission peak appears on the high-energy side of the three PL peaks originating from the ground-state exciton transitions of the three QW's. This high-energy PL peak with an intensity comparable to the one of the other three peaks is located near an excited-subband (n=2) exciton state of the widest QW. By investigating the position dependence of the PL spectra across a 2-inch wafer, which exhibits a decreasing Al mole fraction x from the center to the edge, the PL intensity of the high-energy peak can be correlated with the one of the ground-state in the widest QW. This correlation indicates that this high-energy PL peak is most likely related to the n=2 exciton state of the widest QW because of the energy alignment of the excited subband state relative to the barrier band edge.

PACS: 78.66.Fd, 71.35.-y, 73.21.Fg.
Keywords: photoluminescence, quantum well, excited-subband exciton state

1. Introduction

Population inversion of quantum well (QW) subband states is of key importance for the realization of quantum-cascade lasers [1,2]. In order to improve these devices, it is very important to design them in such a way that the population inversion of the involved subbands is maximized. However, the intersubband relaxation time of the carriers is very fast so that the instantaneous population in the excited subband can only be observed in the picosecond time domain under pulsed photoexcitation [3-5].

In this paper, we report the observation of a surprisingly strong photoluminescence (PL) emission band originating from an excited-subband exciton state in a GaAs/Al$_x$Ga$_{1-x}$As triple QW system with different well thicknesses. Low-temperature steady-state PL experiments show four distinct PL peaks. The three PL peaks on the low-energy side originate from the excitonic transitions of the corresponding ground state in each QW. The PL peak highest in energy is located near an excited-subband exciton state of the widest QW. Its intensity is comparable to the PL intensity of all other peaks. By studying the position, i. e., the Al mole fraction, dependence of the PL spectra across a 2-inch wafer, we find that the intensity variations of the PL line highest in energy appear to be correlated with the one lowest in energy. This indicates that the PL line highest in energy most likely originates from the excited-subband exciton state of the widest QW.

2. Experiment

A nominally undoped sample used in the present study was grown on a 2-inch diameter GaAs(100) substrate by molecular-beam epitaxy (MBE). The growth was performed in a Varian Gen-II machine with a substrate rotation speed of 5 rpm under optimized V/III flux conditions [6]. The sample consists of three GaAs QW's with nominal thicknesses of 7.8, 5.5, and 3.5 nm labeled QW1, QW2, and QW3, respectively, which are separated by 36-nm thick Al$_x$Ga$_{1-x}$As barriers with a nominal Al mole fraction x=0.17. These five layers are sandwiched between a pair of 72-nm Al$_{0.17}$Ga$_{0.83}$As barriers. The complete structure is embedded in a pair of 190-nm (surface side) and 430-nm (substrate side) Al$_{0.3}$Ga$_{0.7}$As cladding layers. According to the growth experience with this MBE machine and different types of pyrolytic BN crucibles used for the Al and Ga cells, the Al flux arrival rate (J_{Al}) slightly decreases from the center to the edge of the wafer, while the Ga flux arrival rate (J_{Ga}) is fairly uniform (deviations of less than 2%)

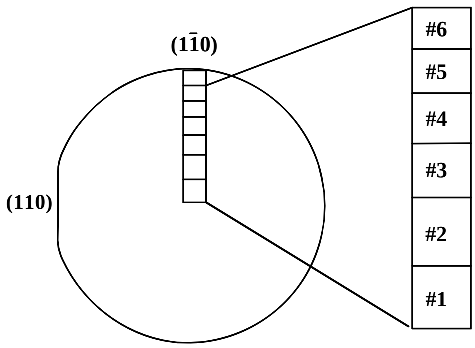

Fig. 1: Position of the six pieces of the GaAs/Al$_x$Ga$_{1-x}$As QW's on the 2-inch wafer numbered from #1 at the center to #6 at the edge.

across the whole 2-inch wafer. Therefore, the Al concentration in the $Al_xGa_{1-x}As$ barriers, which is basically determined by the ratio $J_{Al}/(J_{Al} + J_{Ga})$, is expected to possess a gradient from the center to the edge of the wafer. In order to study the Al mole fraction dependence of the optical properties, the 2-inch wafer was cleaved into six pieces as shown in Fig. 1. The six pieces were numbered from #1 at the center to #6 at the edge as indicated in Fig. 1. All six pieces were mounted in a closed-cycle He cryostat to measure PL and PL excitation (PLE) spectra at 13 K. For the PL measurements, a cw He-Ne laser (λ=632.8 nm) with a power density of about 10 W/cm^2 was used for excitation combined with a standard lock-in system for detection. For the PLE measurements, a halogen lamp dispersed by a 32-cm monochromator was used for excitation.

3. Results and discusion

Figure 2 shows the normalized PL spectra of the pieces #1 to #6 measured at 13 K. The PL spectra of pieces #1 to #5 contain four emission bands. The PL peaks labeled 1, 2, and 3 in Fig. 2 are assigned to the excitonic transitions of the corresponding ground state in QW1, QW2, and QW3, respectively. This assignment is confirmed by a calculation of the energies of the lowest conduction and highest valence subband (heavy holes) including a correction for the exciton binding energy as discussed below. In addition to these ground-state transitions, there is a fourth PL peak at higher energies labeled 4 in Fig. 2. For pieces #1 to #4, it exhibits a similar intensity as peaks 1 to 3. Note that both the PL energy and the relative PL intensity of all observed transitions vary with sample position, i. e., Al mole fraction. In particular, the PL intensity of line 4 first increases from #1 to #2 and then decreases from #2 to #6, exhibiting a maximum value for piece #2. For piece #5, the PL intensity of line 4 is already strongly reduced. The second important point to note is that the PL intensities of line 1 (the excitonic peak of the ground-state in QW1) and line 4 appear to be correlated, i.e., when line 4 becomes weaker with increasing distance from the center of the wafer, the intensity of line 1 is also reduced. This result indicates that line 4 most likely originates from the excited-subband exciton state of QW1.

The variations of the PL spectral characteristics mentioned above can be related to changes in the Al concentration of the nominal $Al_{0.17}Ga_{0.83}As$ barrier. Figure 3 shows a series of PLE spectra for pieces #1 to #6 by setting the PL detection energy at the low-energy tail of the QW3 transition. In these PLE spectra, the exciton absorption peak of the $Al_{0.17}Ga_{0.83}As$ barrier exhibits a red-shift from the center to the edge of the wafer. This systematic shift of the exciton absorption peak of the $Al_{0.17}Ga_{0.83}As$ barrier demonstrates that the Al mole fraction decreases from the center to the edge of the wafer. In addition, the heavy-hole (e1-hh1) and light-hole (e1-lh1) exciton transitions are clearly

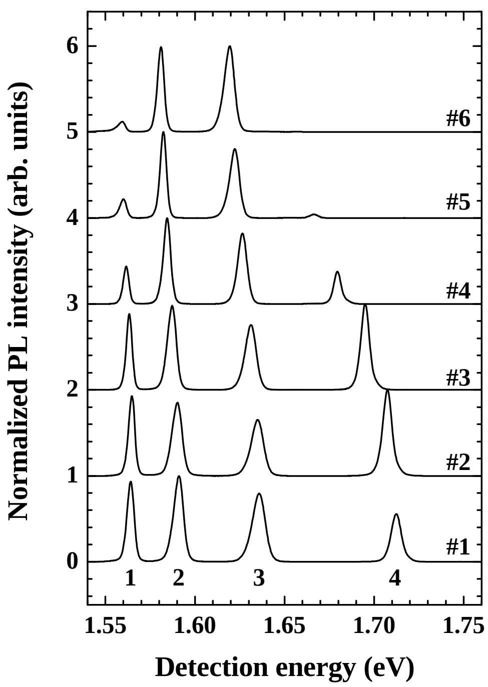

Fig. 2: PL spectra of the pieces #1 to #6. The PL intensity in each spectrum is normalized by the corresponding maximum value. The base lines of the PL spectra are vertically shifted for clarity.

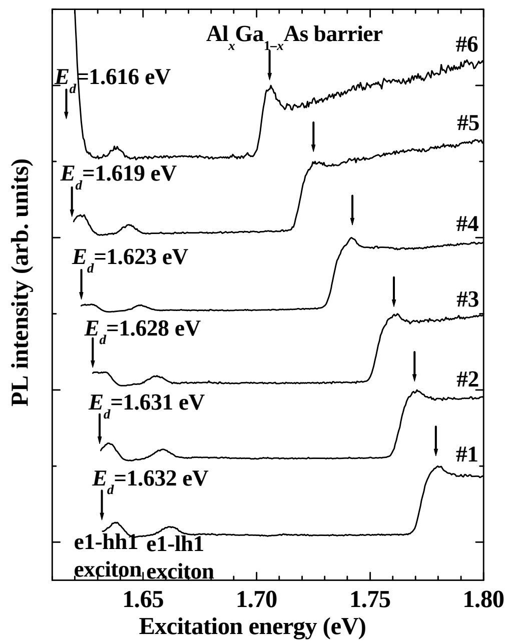

Fig. 3: PLE spectra of the pieces #1 to #6. The PL detection energy E_d as well as the absorption peak of the $Al_xGa_{1-x}As$ barrier is marked for each spectrum by arrows.

observed for QW3, which are red-shifted, when the position on the wafer is moved from the center to the edge. The shifts of the PL as well as the PLE peaks are therefore explained by a decrease of the Al mole fraction in the $Al_{0.17}Ga_{0.83}As$ barrier, which agrees with the growth trend mentioned above. We note that the transition energy of line 4 in Fig. 2 coincides with the high-energy exciton resonance not visible in the PLE spectra of QW2 and QW3, but observable in the PLE spectra of QW1 [7]. This observation confirms the assumption that PL line 4 is not connected with QW2 and QW3, but it is directly related to QW1.

We used the absorption peak of the (Al,Ga)As barrier with a nominal composition of 0.17 from the PLE measurements to determine the Al mole fraction x as shown in Fig. 4 by the open squares. Here we assume that the energy gap E_g of $Al_xGa_{1-x}As$ is given by

$$E_g = E_{\mathrm{GaAs}}(T) + 1.247x \quad [\mathrm{eV}] \qquad (1)$$

and

$$E_{\mathrm{GaAs}}(T) = 1.519 - \frac{5.408\times 10^{-4}T^2}{T+204} \quad [\mathrm{eV}] \qquad (2)$$

where $E_{\mathrm{GaAs}}(T)$ denotes the value of the GaAs energy gap as a function of temperature [8]. The exciton binding energy is assumed to be 8.0 meV for the nominal $Al_{0.17}Ga_{0.83}As$ barrier [9]. The experimental energies of the PL peaks for QW1, QW2, and QW3 are plotted by the solid symbols in Fig. 4. We calculated the transition energies of the ground-state excitonic transition (e1-hh1)

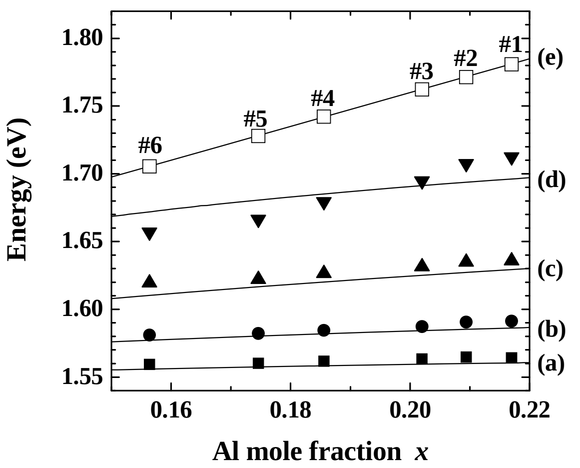

Fig. 4: Exciton peak energy (□) of the $Al_xGa_{1-x}As$ barrier determined from the PLE measurements as a function of Al mole fraction x. Transition energies of the ground exciton states in QW1, QW2, and QW3 and the excited state in QW1, labeled by solid symbols (■, ●, ▲, and ▼, respectively). Solid lines indicate the calculated transition energies of (a) e1-hh1 exciton in QW1, (b) the e1-hh1 exciton in QW2, (c) the e1-hh1 exciton in QW3, (d) the first excited subband (e2-hh2) exciton in QW1, and (e) the exciton in the $Al_xGa_{1-x}As$, respectively.

for QW1, QW2, and QW3, which are indicated by the solid lines in Fig. 4. In the calculations, we use a finite potential well model within the envelope-function approximation [6,10]. Here the band offset ratio for the conduction and valence band is assumed to be 0.64:0.36 and the effective masses are $(0.0665+0.083x)m_0$ for electron and $(0.34+0.41x)m_0$ for heavy-hole, respectively, in $Al_xGa_{1-x}As$. In addition, we took into account the nonparabolicity of the band structure, i. e., an energy-dependent electron effective mass for GaAs [6,10]. Reasonable agreement between theory and experiment is obtained, when we assume an exciton binding energy of the e1-hh1 exciton of 8.0 meV for QW1, of 9.1 meV for QW2, and of 10.9 meV for QW3 [11]. For the PL band on the high-energy side, the transition energy roughly coincides with the first excited (e2-hh2) parity-allowed transition of QW1, also indicated by a solid line in Fig. 4. The exciton binding energy is not taken into account for this excited-subband exciton transition of QW1.

When the Al mole fraction in the barrier is increased, the PL peak of each QW shows a blue-shift due to the increased confinement energy. Comparing the experimental PL peak energies with the calculated transition energies in Fig. 4, it seems that the observed shifts of the PL peak on the high-energy side deviate from the expected values of the transition energy for the excited-state transition in QW1. Nevertheless, it is clearly connected with QW1 because of the correlation of the respective PL intensities with varying Al mole fraction in the barrier.

In two-dimensional QW systems, the emission associated with the first excited (e2-hh2) parity-allowed transition is generally not observed, since the intersubband relaxation time is very fast (typically in the picosecond range) due to longitudinal-optical (LO) phonon scattering [3-5]. The origin of the enhancement of the high-energy PL line may be explained following Ref. [12]. When electron-hole pairs or excitons are generated above the barrier, they can be transferred into the QW's. When the excited-subband exciton state in QW1 is located slightly below the barrier exciton states, preferably by more than the GaAs LO phonon energy, those excitons, which come from the barrier exciton states, can relax into the excited-subband exciton state with a negligibly small in-plane momentum ($k_{\|} \sim 0$), in addition to the usual intrasubband relaxation path within the ground exciton state. Those excitons with small $k_{\|}$ values must vertically relax into the ground states in momentum and real space. However, when the relaxation rate is not efficient because of momentum conversation, an efficient PL signal can be expected from the excited-subband exciton state. In this case, the radiative recombination processes are governed by the exciton population in the excited state. When the Al mole fraction in the barrier is decreased, both the ground- and the excited-state emission decrease in intensity. This observation may be explained by competitive carrier capture processes by QW2 and QW3 [13]. This is also evidenced by the relative increase of the QW2 and QW3 intensities with respect to the QW1 intensity as shown in

Fig. 2.

4. Conclusion

In summary, the PL properties of a GaAs/ $Al_xGa_{1-x}As$ quantum well system containing three QW's with different thicknesses have been studied as a function of Al mole fraction in the $Al_xGa_{1-x}As$ barrier. Four distinct PL peaks have been observed at 13 K. The three PL peaks at lower energies are associated with the exciton transition of the ground-state in each QW. The PL peak highest in energy is located near the excited-subband exciton state of the widest QW and just below the band gap of the $Al_xGa_{1-x}As$ barrier. Furthermore, the PL intensities of the peaks lowest and highest in energy appear to be strongly correlated, indicating that the peak highest in energy most likely originates from the excited-subband exciton state of the widest QW.

References

[1] J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science 264 (1994) 553.

[2] For a review, see: C. Gmachl, F. Capasso, D. L. Sivco, and A. Y. Cho, Rep. Prog. Phys. 64 (2001) 1533.

[3] R. Ferreira and G. Bastard, Phys. Rev. B 40 (1989) 1074.

[4] H. T. Grahn, H. Schneider, W. W. Rühle, K. v. Klitzing, and K. Ploog, Phys. Rev. Lett. 64 (1990) 2426.

[5] Bipul Pal and A. S. Vengurlekar, Appl. Phys. Lett. 79 (2001) 72.

[6] K. Fujiwara, H. Katahama, K. Kanamoto, R. Cingolani, and K. Ploog, Phys. Rev. B 43 (1991) 13978.

[7] K. Fujiwara, H. T. Grahn, L. Schrottke, and K. H. Ploog, Proc. 25th Int. Conf. Phys. Semicond. (Springer-Verlag, Berlin, 2001) 627

[8] H. C. Casey and M. B. Panish, in: Heterostructure Lasers (Academic, New York, 1978), Pt. A.

[9] P. J. Pearah, W. T. Masselink, J. Klem, T. Henderson, H. Morkoc, C. W. Litton, and D. C. Reynolds, Phys. Rev. B 32 (1985) 3857.

[10] H. T. Grahn (Ed.), Semiconductor Superlattices: Growth and Electronic Properties, (World Scientific, Singapore, 1995).

[11] L. C. Andreani and A. Pasquarello, Phys. Rev. B 42 (1990) 8928.

[12] K. Fujiwara, H. T. Grahn, L. Schrottke, and K. H. Ploog, Physica E 11 (2001) 215

[13] K. Fujiwara, M. Ohe, M. Matsuo, T. Nogami, H. T. Grahn, and K. H. Ploog, Inst. Phys. Conf. Ser. No 166 (IOP, Bristol, 2000), p. 103.