Technical University of Denmark

Stokes and anti-Stokes photoluminescence towards five different In-x(Al0.17Ga0.83)(1-x)As/Al0.17Ga0.83As quantum wells

Machida, S.; Tadakuma, T.; Satake, A.; Fujiwara, K.; Folkenberg, J. R.; Hvam, Jørn Marcher

Published in:
Journal of Applied Physics

Link to article, DOI:
10.1063/1.2121928

Publication date:
2005

Document Version
Publisher's PDF, also known as Version of record

Stokes and anti-Stokes photoluminescence towards five different $In_x(Al_{0.17}Ga_{0.83})_{1-x}As/Al_{0.17}Ga_{0.83}As$ quantum wells

S. Machida, T. Tadakuma, A. Satake, and K. Fujiwara
Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan

J. R. Folkenberg
Crystal Fibre A/S, Blokken 84, DK-3460 Birkerød, Denmark

J. M. Hvam
Research Center COM, Technical University of Denmark, Building 349, DK-2800 Kgs. Lyngby, Denmark

(Received 11 May 2005; accepted 15 September 2005; published online 31 October 2005)

Stokes and anti-Stokes photoluminescence (AS-PL) has been investigated in a step-graded $In_x(Al_{0.17}Ga_{0.83})_{1-x}As/Al_{0.17}Ga_{0.83}As$ quantum-well (QW) heterostructure consisting of five QWs with different x values. Stokes PL spectra of this sample show a significant difference in PL intensity between the wells under indirect excitation conditions due to the existence of competitive resonant and nonresonant capture processes, while they exhibit a rather uniform PL intensity distribution under direct excitation. When the excitation wavelength is tuned to 810 nm for AS-PL detection, it is transparent to the five QWs and thus the photoabsorption can only occur in the GaAs (rear buffer and front cap) layers. It is found that the AS-PL spectra show a similar intensity distribution to the one observed under the indirect excitation. This result means that the AS-PL intensity distribution of the QWs is basically determined by the competitive capture of photoexcited carriers through the thick barriers, generated far from the five wells due to the nonlinear excitation processes in GaAs.
 [DOI: 10.1063/1.2121928]

I. INTRODUCTION

Anti-Stokes photoluminescence (AS-PL) where output emission energy is larger than input photon energy has been received recent attention in a variety of quantum-well (QW) heterostructures.[1–3] In most of the previous studies asymmetric double QWs with higher and lower subband exciton states are excited resonantly to the lower exciton energy state and the higher exciton emission is observed, owing to the nonlinear excitation processes such as Auger excitation[4] as well as two-step two-photon absorption (TS-TPA) processes.[1,2,5] In the asymmetric double QW systems the high-energy radiative recombination can always occur next to the low-energy QW region. Therefore, carrier transfer from the photoexcitation region to the recombination place can occur within a short distance.

In this paper, PL up-conversion has been investigated in a unique QW system comprising of five QWs with spectrally discriminative exciton bands. PL intensity distribution of this sample shows a significant difference between direct and indirect excitation conditions. That is, under indirect barrier excitation conditions a strongly nonuniform PL intensity distribution is observed between the five different wells, while no such difference in PL intensity is found by direct well excitation. Thus, PL enhancement and reduction observed for the different wells by indirect excitation are attributed to the competitive capture of carriers from the barrier into the five wells. When the excitation wavelength is tuned to 810 nm for AS-PL observation by spatially selective photoexcitation of the GaAs layers, AS-PL signals for the QWs are clearly detected whose intensity distribution is very similar to that of Stokes-shifted PL of five wells under indirect photoexcitation. That is, the AS-PL intensity of the QWs is basically determined by the competitive capture of photoexcited carriers through the thick barriers.

II. EXPERIMENTAL

The sample used for this study was grown by molecular beam epitaxy on a GaAs (100) substrate at 530 °C. After a 200 nm thick GaAs buffer layer on the substrate, the step-graded $In_x(Al_{0.17}Ga_{0.83})_{1-x}As/Al_{0.17}Ga_{0.83}As$ QW structure consisting of five QWs with different x values was grown. The QW structure is embedded in a pair of 200 nm thick $Al_{0.17}Ga_{0.83}As$ barrier layers. The In mole fraction is 5.3%, 8.8%, 12%, 15%, and 18% in QW1, 2, 3, 4, and 5, respectively, from the substrate side.[6] Each well is electronically isolated by 30 nm thick $Al_{0.17}Ga_{0.83}As$ barrier layers. The growth is terminated by a 100 nm thick GaAs cap layer. PL spectra were measured at 14-15 K in a closed-cycle He cryostat using a He-Ne laser (632.8 nm and 10 W/cm^2). A Jobin-Yvon (HR320) monochromator and a computer-controlled digital lock-in-amplifier system were used for the PL detection. For PL excitation (PLE) spectra, a combination of a 300 W halogen lamp and a monochromator was used for wavelength tunable and weak ($\sim$10 μW) excitation. A wavelength tunable Ti-sapphire laser (710-810 nm with a maximum power density of 500 W/cm^2 in a quasi-cw mode) was used for AS-PL experiments.

III. RESULTS AND DISCUSSION

Figure 1 shows a PL spectrum of the sample at 14 K when a He-Ne laser was used as an excitation source (indirect excitation). The PL spectrum shows five emission bands

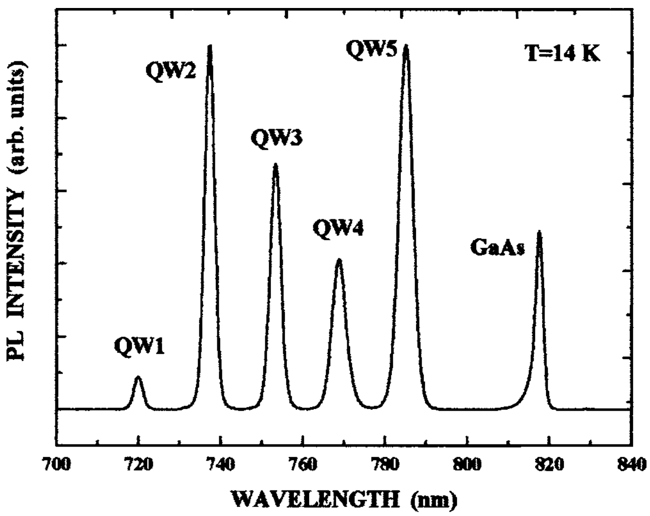

FIG. 1. PL spectrum measured at 14 K using a He-Ne laser (632.8 nm wavelength and a 10 W/cm^2 power density) for indirect excitation.

at 720, 737, 753, 769, and 785 nm, which we attribute to the five QW layers, as assigned in Fig. 1.[6] A PL peak observed at 817 nm is ascribed probably to bound excitons in the GaAs layers. Under the indirect excitation at 632.8 nm where incident photons are basically absorbed in the front 200 nm thick $Al_{0.17}Ga_{0.83}As$ barrier layer, the PL intensity is not uniformly distributed for the five emission bands. Some of the PL bands such as QW2, QW3, and QW5 exhibit higher intensities than those of QW1 and QW4 in agreement with our previous studies.[7,8]

To investigate the excitation wavelength dependence of the PL intensity distribution, PL spectra are measured using a halogen lamp/monochromator system for wavelength tunable and weak (~10 μW) excitation. When excited above the barrier band gap by excitation wavelength at 633 nm (indicated by a solid curve for indirect excitation), as shown in Fig. 2, the five PL bands show a similar intensity distribution to the one in Fig. 1 exhibiting the reduced PL for QW4 and

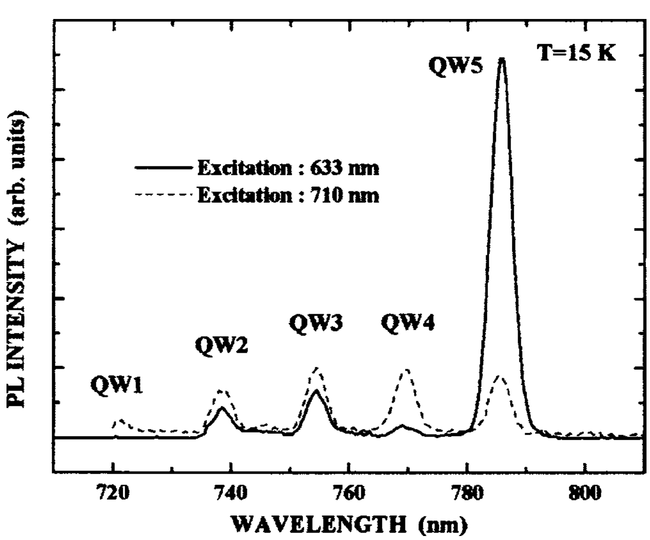

FIG. 2. PL spectra under direct (dashed curve) and indirect (solid curve) excitation conditions using a halogen lamp for weak photoexcitation at 15 K. Note a significant difference in PL intensity between the five wells and between the two excitation conditions.

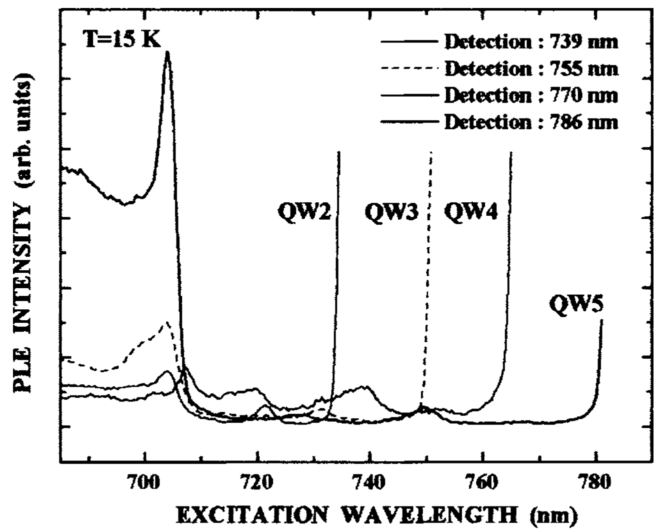

FIG. 3. PLE spectra of QW2, 3, 4, and 5 at 15 K with the detection wavelength set at 739, 755, 770, and 786 nm, respectively. When the excitation wavelength is tuned at the barrier band edge (705 nm), PL intensities are drastically changed among the different wells. Note a reduction of the PLE intensity for QW4 at 705 nm.

QW1, except for the strongest PL signal for QW5. This PL enhancement observed only for QW5 under the very weak excitation conditions can be explained by the efficient capture of photogenerated carriers from the front $Al_{0.17}Ga_{0.83}As$ barrier layer into the QW5, since the incident excitation light is mostly absorbed by the front barrier layer and not effectively penetrating into the bottom barriers. On the other hand, when only the five QWs are directly and selectively excited at 710 nm wavelength (indicated by a dashed curve for direct excitation), a rather uniform PL intensity distribution is obtained. That is, the PL intensity distributions are quite different between the two (direct and indirect) excitation conditions. The weak PL intensity observed for QW1 in Fig. 2 may be due to the phonon-assisted carrier escape towards the other wells even at 15 K because of the shallowest potential depth.[7,8] Note that the weak PL intensity for QW4 under the indirect excitation is significantly intensified by the direct excitation in Fig. 2. Therefore, these PL results suggest that the nonuniform PL intensity distribution under the indirect excitation is determined by the competitive resonant capture processes from the barrier layer into the different QWs.

In order to directly verify how efficiently the carrier capture probability from the barriers can be changed between the different wells, a series of PLE spectra covering the barrier band edge region are displayed in Fig. 3 by setting the PL detection wavelength at the four PL bands of QW2, 3, 4, and 5. In these spectra of Fig. 3, PLE peaks due to the light-hole exciton resonances are seen near 722 nm (QW2), 732 nm (QW3), 740 nm (QW4), and 751 nm (QW5) in agreement with Ref. 6. When the excitation wavelength is tuned at the barrier band edge around 705 nm, the PL intensity of QW3 and QW5 is drastically enhanced due to the resonant capture,[9] being consistent with the PL results of Figs. 1 and 2, especially under the weak excitation power. On the other hand, the PL intensity of QW4 is significantly reduced at the

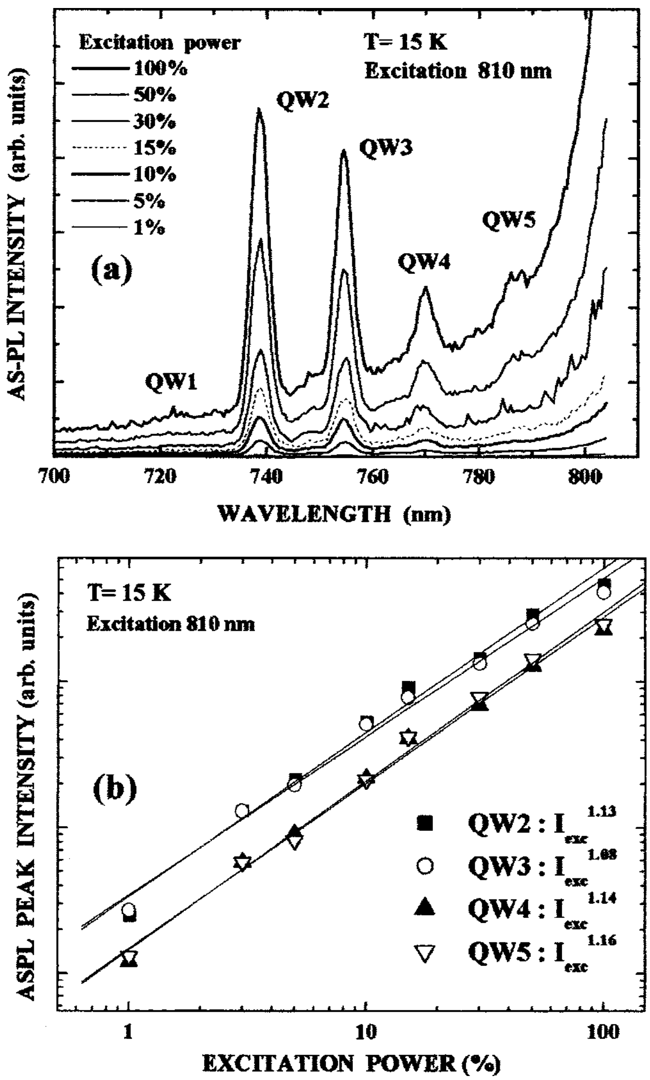

FIG. 4. (a) Anti-Stokes PL spectra measured at 15 K using an excitation wavelength of 810 nm with varying the excitation power (the 100% maximum power density is 500 W/cm^2). (b) Anti-Stokes PL intensity for QW2, QW3, QW4, and QW5 as a function of excitation power (points are experimental data and solid lines for fitting).

wavelength resonant to the barrier exciton in Fig. 3 after the seeming PLE peak around 710 nm. This result is surprising since the less efficient PL intensity of QW4 under the indirect excitation is obtained than the direct excitation. This is due to the two facts that absorbed photons by the QW4 layer decrease by the indirect excitation at 705 nm because of the strong barrier resonant absorption (mainly by the front barrier layer) and that the photogenerated carriers are mostly captured by the other competitive QW3 and QW5 layers where the resonant capture processes are operative. These PLE results directly prove that the capture processes from the barriers by the five different wells are in fact different due to the barrier resonant and nonresonant excitation to the excited subband states in the wells.[9]

Figure 4(a) shows AS-PL spectra for the five QWs at 15 K when the GaAs bulk layer is excited at 810 nm with varying the excitation power. Note that by the excitation at 810 nm the QWs are all transparent and the AS-PL signals whose energy is larger than the energy of incident photons are only possible by nonlinear excitation processes such as TS-TPA. In fact, the AS-PL intensity ($I_{\text{AS-PL}}$) as a function of excitation power (I_{exc}) shows a nonlinear power dependence [see Fig. 4(b)]. That is, the $I_{\text{AS-PL}}$ is proportional to $I_{\text{exc}}^{1.13}$ (QW2), $I_{\text{exc}}^{1.08}$ (QW3), $I_{\text{exc}}^{1.14}$ (QW4), and $I_{\text{exc}}^{1.16}$ (QW5). We also note that the observed AS-PL intensity distribution among the five QWs is very similar to the PL intensity distribution observed under the indirect excitation above the $Al_{0.17}Ga_{0.83}As$ barrier band gap (see Figs. 1 and 2). Therefore, the AS-PL intensity distribution resembling to those for Figs. 1 and 2 under the indirect excitation is due to the photoexcitation in the GaAs layers and the resultant carrier transfer via the $Al_{0.17}Ga_{0.83}As$ barrier layers (indirect transfer). In this excitation wavelength condition, it is transparent to the five QWs and thus the photoabsorption can only occur in the front cap and rear buffer GaAs layers. Assuming a value of 8×10^3 cm^{-1} for the GaAs optical absorption coefficient,[10] it is estimated that 8% of the incident photons are absorbed in the front GaAs layer and a majority of incoming photons are absorbed in the bottom GaAs layer. We thus attribute the relatively smaller AS-PL intensity for QW5 in Fig. 4 to the fact that the carriers are not effectively captured by QW5 since they must traverse over the QW1-4 layers to reach the topmost QW5 from the bottom GaAs layer after the nonlinear excitation processes. This reduction of the AS-PL intensity for QW5 is in sharp contrast to the PL enhancement for QW5 in Fig. 2 under the indirect and weak excitation where the photoexcited carriers are mostly generated in the front $Al_{0.17}Ga_{0.83}As$ barrier layer nearest to the QW5. Furthermore, while the PL intensity for QW2 is weak for the weak indirect excitation as seen in Fig. 2 (due to the front $Al_{0.17}Ga_{0.83}As$ barrier excitation), the strongest AS-PL intensity observed for QW2 in Fig. 4(a) is due to the indirect transfer of photoexcited carriers from the bottom GaAs layer. These results mean that the AS-PL intensity distribution from the five QWs is strongly influenced by the position where the photoexcited carriers are generated even in the PL up-conversion processes.

IV. CONCLUSION

In summary, AS-PL properties have been investigated in a unique step-graded $In_x(Al_{0.17}Ga_{0.83})_{1-x}As/Al_{0.17}Ga_{0.83}As$ QW structure consisting of five QWs with different x values. The Stokes PL intensity distribution due to the five QWs shows a significant difference under direct and indirect excitation conditions. When the excitation wavelength is tuned to 810 nm for exciting the GaAs layers, AS-PL signals from QW1-5 are detected via nonlinear excitation processes. The AS-PL intensity distribution between the five QWs is very similar to that for the Stokes PL intensity under indirect excitation. This result means that the AS-PL intensity distribution of the QW1-5 is basically determined by the competitive capture of photoexcited carriers through the thick barriers, generated far from the five wells due to the nonlinear excitation processes in GaAs.

[1]R. Hellmann, A. Euteneuer, S. G. Hense, J. Feldmann, P. Thomas, E. O. Gobel, D. R. Yakovlev, A. Wagg, and G. Landwehr, Phys. Rev. B **51**, 18053 (1995).
[2]H. M. Cheong, B. Fluegel, M. C. Hanna, and A. Mascarenhas, Phys. Rev.

B **58**, R4254 (1998).
[3]S. C. Hohng, D. W. Khang, Y. H. Ahn, J. Y. Lee, S. Y. Kihm, D. H. Kim, W. S. Kim, J. C. Woo, D. S. Kim, D. S. Citrin, D. H. Woo, E. K. Kim, S. H. Kim, and K. S. Lim, Phys. Rev. B **60**, 8883 (1999).
[4]W. Seidel, A. Tivkov, J. P. André, P. Voisin, and M. Voos, Phys. Rev. Lett. **73**, 2356 (1994).
[5]Y.-H. Cho, D. S. Kim, B.-D. Choe, H. Lim, J. I. Lee, and D. Kim, Phys. Rev. B **56**, R4375 (1997).
[6]J. R. Jensen, J. M. Hvam, and W. Langbein, J. Appl. Phys. **86**, 2584 (1999).
[7]S. Machida, M. Matsuo, K. Fujiwara, J. R. Jensen, and J. M. Hvam, Physica E (Amsterdam) **13**, 182 (2002).
[8]S. Machida, M. Matsuo, K. Fujiwara, J. R. Folkenberg, and J. M. Hvam, Phys. Rev. B **67**, 205322 (2003).
[9]A. Fujiwara, Y. Takahashi, S. Fukatsu, Y. Shiraki, and R. Ito, Phys. Rev. B **51**, 2291 (1995).
[10]Y. Masumoto, M. Matsuura, S. Tarucha, and H. Okamoto, Phys. Rev. B **32**, 4275 (1985).