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# Anti-Stokes photoluminescence between $In_x(Al_{0.17}Ga_{0.83})_{1-x}As$/ $Al_{0.17}Ga_{0.83}As$ quantum wells with different $x$ values

S. Machida, T. Tadakuma, and K. Fujiwara

Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan

## Abstract

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) system consisting of five QWs with different $x$ values. When a low-energy heavy-hole (1s) exciton state in a particular well is resonantly photoexcited, the high-energy heavy-hole (1s) exciton in the nearest-neighbor well shows a stronger AS-PL intensity than other QWs beyond the nearest-neighbor QW. The AS-PL intensity of (1s) excitons observed in each well shows a drastic position dependence on where carriers are resonantly photoexcited, indicating energy transfer processes with a spatial position dependence. These results mean that the up-conversion phenomena responsible for generating high-energy carriers can be influenced by transfer and capture processes into the high-energy exciton state in addition to nonlinear excitation mechanisms.

Keywords: Quantum well, InAlGaAs, Photoluminescence up-conversion, Nonlinear optical properties

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## 1. 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 [1-9]. Nonlinear excitation mechanisms such as two-step two-photon absorption (TS-TPA) including photon recycling, Auger excitation processes, and exciton-dipole interactions have been proposed to explain the high-energy photon generation. In most of the previous studies, however, asymmetric double QWs with higher and lower subband (1s) exciton states are excited resonantly to the lower energy heavy-hole (1s) exciton state in the wider well and the higher energy exciton emission is observed (as schematically shown in Fig. 1), as a result of the nonlinear excitation processes within or near the low-energy exciton state including the barrier/well interface regions. In the asymmetric double QW systems the radiative recombination in the high-energy (1s) exciton state 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, AS-PL emissions due to nonlinear up-conversion processes have been investigated in a unique QW system where there are a number of high-energy exciton states for radiative recombination in QW regions with spectrally discriminative emission bands. That is, the AS-PL emission can occur in a sequence of different QWs spatially well-separated from the low-energy region for excitation. Thus, the emission region for high-energy photons may be located at the nearest-neighbor well and further beyond the nearest-neighbor QW from the low-energy region where the nonlinear excitation processes should occur. Therefore, position-dependence of the anti-Stokes energy transfer may be studied. Depending on the photoexcitation wavelength selected to resonantly excite the particular QW or to excite only the bulk GaAs region far from the QW layers, AS-PL emissions from the QWs show significantly different intensities each other. Influences on the photoexcitation energy transfer between the QWs have been discussed by studying the photoexcitation position dependence using resonant excitation.

## 2. Experimental

The sample used for the present study consists of five $In_x(Al_{0.17}Ga_{0.83})_{1-x}As$ quantum wells, grown by molecular beam epitaxy on a GaAs (100) substrate [10]. After a 200 nm thick GaAs buffer layer on the substrate, the stepped $In_x(Al_{0.17}Ga_{0.83})_{1-x}As/Al_{0.17}Ga_{0.83}As$ 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. Nominal thicknesses of the five wells are all about 8 nm. 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 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 PL detection. A wavelength tunable Ti-sapphire laser (710-810 nm with a maximum (100%) power density of 500 W/cm$^2$ in a quasi-cw mode) was also used for PL and AS-PL experiments. An excitation density $n_{exc}$ of electron/hole pairs at 500 W/cm$^2$ is estimated to be ~$10^{12}$ cm$^{-2}$. Variable excitation power was controlled by neutral density filters.

## 3. Results and discussion

Figures 2(a) and 2(b) show PL spectra of the QW sample measured under indirect (excitation wavelength of 632.8 nm) and direct (excitation wavelength of 715 nm) excitation, respectively. As already discussed in our previous studies [11-13], the PL intensity distribution of this sample shows distinct five emission bands (assignment is given in Fig. 2(a)). They exhibit a significant difference in intensity between direct and indirect excitation conditions. That is, under indirect barrier excitation conditions a strongly non-uniform PL intensity distribution is observed between the 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 [14]. This is because of the resonant capture of the photoexcited carriers, as easily verified by PL excitation (PLE) spectra [13], in strong contrast to the case of direct excitation in (b) where the PL intensity of the five QWs shows rather uniform intensity distribution except for the weak QW1, probably due to a carrier thermal escape even at 15 K [12]. Figure 2(c) shows an AS-PL spectrum when only the GaAs bulk layer is excited at 810 nm. Note that by excitation at 810 nm the QWs are all transparent and the AS-PL intensity in each well is originated from the nonlinear photoexcitation in GaAs to generate high-energy carriers and resultant carrier transfer via the thick $Al_{0.17}Ga_{0.83}As$ barrier layers (indirect transfer) [13]. Thus, their AS-PL intensities resemble to those of PL in Fig. 2(a) taken under indirect barrier photoexcitation except for QW5. That is, the AS-PL intensity of the QWs is basically determined by the competitive capture of photoexcited carriers through the thick barriers. Assuming a value of $8\times10^{3}$ $cm^{-1}$ for the GaAs optical absorption coefficient [15] at 810 nm, 8 % of the incident photons are absorbed in the front GaAs cap 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. 2(c) to the less efficient carrier transfer because the carriers must traverse over the QW1-4 layers to reach the topmost QW5 from the bottom GaAs layer after the nonlinear excitation processes.

Figure 3 shows three AS-PL spectra for QW1-4 when the excitation wavelength is tuned to 780, 785, and 790 nm. The excitation wavelength of 785 nm is close to resonant to the heavy-hole (1s) exciton state in the QW5 layer. We note that the AS-PL intensity for QW4 is significantly enhanced with the 785 nm excitation, but with using longer and shorter excitation wavelengths at 790 and 780 nm the AS-PL intensity is much reduced. This fact rules out a possibility of the AS-PL intensity caused by the scattered laser light. The enhanced AS-PL intensity for QW4, when the neighboring QW5 with the lower exciton energy is resonantly photoexcited, is in strong contrast to the reduced AS-PL intensity observed for QW4 in Fig. 2(c) by the indirect energy transfer. This is because of the difference in photon energy used for the low-energy photoexcitation to generate high-energy carriers in the different QW and bulk regions. It is clear from comparison with the two AS-PL spectra that the AS-PL intensity for QW4 is strongly suppressed in Fig. 2(c) due to the competitive capture through the thick barriers, but it is enhanced in Fig. 3 when the up-converted carriers are efficiently captured from the nearest-neighbor well (QW5). This means that the AS-PL intensity distribution coming from the five emission bands strongly depends on the place where the up-converted carriers are generated. It seems that the AS-PL intensity of QW2 is less sensitive to the changes of the excitation wavelength in Fig. 3. As demonstrated in Fig. 2(c), the AS-PL intensity of QW2 is mainly determined by the nonlinear excitation process in the buffer and cap GaAs layers due to the large absorption thickness. Therefore, the insensitivity of the AS-PL intensity of QW2 to the excitation wavelength can be explained, while the AS-PL intensity of QW4 is certainly sensitive to the excitation wavelength leading to the enhancement by the resonance absorption in the nearest-neighbor QW5 layer.

Figure 4 shows an AS-PL spectrum when the excitation is made with the highest power at the 784.4 nm wavelength resonant to the (1s) exciton state in the QW5 layer. A surprisingly strong AS-PL intensity is seen for QW4 when the neighboring QW5 is resonantly photoexcited at $n_{exc}$ of ~$10^{12}$ cm$^{-2}$. Note that the AS-PL intensity from QW1, 2, and 3 is much weaker than QW4. In order to examine the nonlinear photoexcitation processes we have investigated the excitation power dependence on the AS-PL intensity of QW4 for 784.4 nm excitation. It is found that the AS-PL intensity ($I_{AS-PL}$) of QW4 as a function of excitation power ($I_{ex}$) reveals $I_{ex}^{1.37}$ dependence reflecting the nonlinear excitation processes in QW5, while that of QW3 (QW2) shows $I_{ex}^{1.04}$ ($I_{ex}^{1.02}$) dependence. This result means that the AS-PL signal intensity in the system of five QW layers is strongly influenced by the distance between the photoexcitation and the radiative recombination positions. That is, it depends on how far the photoexcited carriers are generated by nonlinear excitation processes in the excitation region relative to the recombination place and how they are transferred to the recombination positions. Figure 5 shows similar results when the excitation is made with the highest power at the 769.5 nm wavelength resonant to the (1s) exciton state in the QW4 layer. It is evident again that the AS-PL intensity for QW3 is strongly enhanced when the neighboring QW4 is resonantly photoexcited. The AS-PL intensity from QW1 and QW2 is found to be weaker. In this case it is estimated that the AS-PL intensity ($I_{AS-PL}$) of QW3 as a function of excitation power ($I_{ex}$) reveals $I_{ex}^{1.33}$ dependence, while that of QW2 shows $I_{ex}^{1.21}$ dependence. Similar to the previous case of the resonant excitation to QW5, the AS-PL intensity distribution shows the significant enhancement for the nearest-neighboring QW3 when the QW4 with the low-energy (1s) exciton state is resonantly photoexcited.

The experimental results shown in Figs. 4 and 5 indicate that the AS-PL signals observed for the different QWs are strongly influenced by the excitation position, and depend on how far the photoexcited carriers are generated by nonlinear excitation processes in the excitation region relative to the recombination place and how they are transferred to the recombination positions. Previously, Viattieri et al. [2] studied the AS-PL signals in the four GaAs/$Al_{0.28}Ga_{0.72}As$ QWs with different well thicknesses of 6, 9, 12, and 18 nm separated by 30 nm thick barriers. When the 18 nm QW was resonantly excited, the AS-PL intensity from the 6 and 9 nm QWs did not show any big difference, in agreement with our result in Fig. 4, since they are not the nearest -neighbor well. However, the AS-PL signal with the detection wavelength set for the 6 nm QW was not enhanced in their AS-PLE spectrum when the nearest-neighbor 9 nm QW is excited at $n_{exc}$ of ~$10^8$ $cm^{-2}$. This difference may be due to the different excitation density of carriers used. In fact, when decreasing the carrier density (to $n_{exc}$ of ~$10^{10}$ $cm^{-2}$) by two orders of magnitude in the inset of Fig. 4, no such big difference in the AS-PL intensity between QW4 and QW3 can be seen.

Schematic potential energy diagrams for the long-range indirect energy transfer corresponding to Fig. 2(c) and for the short-range transfer corresponding to Figs. 4 and 5 are illustrated in Figs. 6(a) and 6(b), respectively. In the case (a) where the photogeneration is made far from the recombination sites the excited carriers must travel through the thicker barriers after the nonlinear excitation. Therefore, the carrier capture probability through the barriers into the individual QWs determines the AS-PL intensity. The particular QW that can efficiently capture the high-energy carriers thus shows the highest AS-PL intensity. The QW2 corresponds to this case in our experiments. In contrast, when the nonlinear excitation place is spatially selected by tuning the resonant photoexcitation wavelength, the AS-PL intensity shows a strong preference to the nearest-neighboring QW. These results mean that the AS-PL intensity is strongly influenced by the nonlinear excitation mechanism itself but also by the real-space carrier transfer and capture processes. That is, it depends on the nonlinear excitation place relative to the recombination site because the photogenerated high-energy carriers must traverse to produce the complex AS-PL intensity distribution.

## 4. 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 AS-PL intensity distribution shows a striking difference between the five QWs, depending on the photoexcitation wavelength selected for nonlinear up-conversion processes. AS-PL intensity observed for the different QWs shows dramatic dependence on where the high-energy carriers are resonantly photoexcited, indicating spatially non-uniform transfer processes with a spatial position dependence.

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## References

[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] A. Viattieri, F. Bogani, A. Miotto, and S. Ceccherini, pys. Stat. Sol. (a)164, 101 (1997).

[3] Y. -H. Cho, D. S. Kim, B. -D. Choe, H. Lim, J. I. Lee, and D. Kim, Phys. Rev. B **56**, R4375 (1997).

[4] H. M. Cheong, B. Fluegel, M. C. Hanna, and A. Mascarenhas, Phys. Rev. B **58**, R4254 (1998).

[5] 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).

[6] T. Kita, T. Nishino, C. Geng, F. Scholz, and H. Schweizer, Phys. Rev. B59, 15358 (1999).

[7] M. Potemski, R. Stepniewski, J. C. Mann, G. Martinez, P. Wyder, and B. Etienne, Phys. Rev. Lett. **66**, 2239 (1991)

[8] W. Seidel, A. Tivkov, J. P. André, P. Voisin, and M. Voos, Phys. Rev. Lett. **73**, 2356 (1994).

[9] A. Tomita, J. Shah, and R. S. Knox, Phys. Rev. B53, 10793 (1996).

[10] J. R. Jensen, J. M. Hvam, and W. Langbein, J. Appl. Phys. **86**, 2584 (1999).

[11] S. Machida, M. Matsuo, K. Fujiwara, J. R. Jensen, and J. M. Hvam, Physica E **13**, 182 (2002).

[12] S. Machida, M. Matsuo, K. Fujiwara, J. R. Folkenberg, and J. M. Hvam, Phys. Rev. B **67**, 205322 (2003).

[13] S. Machida, T. Tadakuma, A. Satake, K. Fujiwara, J. R. Folkenberg, and J. M. Hvam, J. Appl. Phys. 98, 083527 (2005).

[14] A. Fujiwara, Y. Takahashi, S. Fukatsu, Y. Shiraki, and R. Ito, Phys. Rev. B **51**, 2291 (1995).

[15] Y. Masumoto, M. Matsuura, S. Tarucha, and H. Okamoto, Phys. Rev. B **32**, 4275 (1985).

**Captions to figures**

Fig. 1

(a) Anti-Stokes PL (AS-PL) generated by nonlinear excitation processes is detected at the higher energy side than the excitation laser, while Stokes PL is observed at the lower energy side. (b) AS-PL signal generation due to two-step two-photon-absorption (TS-TPA) where photoexcited carriers generated in the low-energy state by the low-energy laser excitation are up-converted by the laser and/or PL to the high-energy state.

Fig. 2

PL spectra measured at 14-15 K under (a) indirect, (b) direct and (c) below QW bandgap excitation conditions. For indirect excitation (a) a He-Ne laser was used at 632.8 nm with a power density of 10 W/cm$^2$, while for direct excitation (b) a Ti-sapphire laser was used at 715 nm with a power density of 500 W/cm$^2$. For the anti-Stokes PL spectrum measured at 15 K an excitation wavelength of 810 nm was used to generate high-energy PL signals in a 710-795 nm range. PL spectra are shifted vertically for clarity.

Fig. 3

Anti-Stokes PL spectra recorded at 15 K using a Ti-sapphire laser tuned at 780, 785, and 790 nm. Note that the anti-Stokes PL intensity for QW4 is higher with the excitation wavelength at 785 nm (nearly resonant to the QW5 exciton state) than those at 780 and 790 nm excitation.

Fig. 4

Anti-Stokes PL spectrum measured at 15 K using an excitation wavelength of 784.4 nm (nearly resonant to the QW5 exciton state) with a power density of 500 W/cm$^2$. The inset figure shows the anti-Stokes PL intensity for QW2, QW3, and QW4 as a function of excitation power (points are experimental data and solid lines for fitting).

Fig. 5

Anti-Stokes PL spectrum measured at 15 K using an excitation wavelength of 769.5 nm (nearly resonant to the QW4 exciton state) with a power density of 500 W/cm$^2$. The inset figure shows the anti-Stokes PL intensity for QW2 and QW3 as a function of excitation power (points are experimental data and solid lines for fitting).

Fig. 6

Schematic models for anti-Stokes energy transfer: (a) long-range transfer to QW1-5 through the barriers from the GaAs buffer region where the nonlinear excitation can occur to generate high-energy carriers, (b) short-range transfer to QW3 and QW4 from the QW5 region where high-energy carriers are resonantly excited by nonlinear processes in QW5.