Time-resolved photoluminescence of a triple GaAs quantum well with growth islands under resonant photoexcitation into the ground and excited states

K. Fujiwara[a]*, H. T. Grahn[b], L. Schrottke[b], and K. H. Ploog[b]

[a]*Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan*

[b]*Paul Drude Institute for Solid State Electronics,*

Hausvogteiplatz 5-7, 10117 Berlin, Germany

Abstract

Exciton radiative recombination properties of an electronically isolated triple GaAs quantum well (QW) with different thicknesses prepared by growth-interrupted molecular beam epitaxy have been investigated with respect to the effect of growth islands present in each QW. When cold excitons with a negligible center-of-mass motion are resonantly photoexcited within each dominant island terrace, the PL dynamics of the split exciton lines reflect the exciton intra-well transfer towards the wider-well terraces. However, dramatic changes are observed in both the PL intensity distribution and the dynamics between the QW's as well as between the island terraces under resonant excitation into an excited (n = 2) state of the widest well located just below the barrier band edge. We tentatively attribute the origin of these changes to the different exciton relaxation pathways in terms of center-of-mass kinetic motion of the two-dimensional excitons.

PACS codes: 78.66.Fd; 78.47.+p; 78.55.Cr; 73.20.Dx

Keywords: Semiconductor quantum well; Photoluminescence; Exciton; Relaxation dynamics

Corresponding author. Telefax: +81-93-884-3221;

E-mail address: fujiwara@ele.kyutech.ac.jp (K. Fujiwara)

1. Introduction

In a system consisting of several quantum wells (QW's) with different thicknesses separated by sufficiently thick barriers, confined electron states in each well are considered to be independent. Therefore, the relevant photoluminescence (PL) properties should exhibit their own characteristics without any correlation between the QW's. However, in recent studies of electronically isolated QW's, unusual PL correlations [1, 2] as well as Stokes and anti-Stokes transfers [3, 4] between the QW excitonic bands have been reported. This observation suggests that the different wells are by no means independent with respect to excitonic radiative recombination and carrier capture/relaxation processes.

In this paper, we investigate the excitonic PL spectral lineshape, the intensity distribution, and the dynamics in a GaAs triple QW with different thicknesses using time-resolved PL (TR-PL) experiments. Pulsed photoexcitation resonantly into the ground exciton states or the first (n=2) excited state of the widest well located just below the barrier band edge results in a drastic change of both, the PL intensity distribution between the QW's and the PL time trace of each QW. These interesting observations suggest that the exciton energy relaxation processes in the widest well are very different from the ones in the other, narrower QW's.

2. Experimental

A triple QW sample was grown at 670 °C on a GaAs (100) substrate by molecular beam epitaxy. Nominal widths of the undoped GaAs QW's are L_Z = 7.8 nm (QW1), 5.5 nm (QW2) and 3.5 nm (QW3) starting from the substrate side, which are electronically isolated by 36 nm thick $Al_{0.17}Ga_{0.83}As$ barriers. These QW layers were grown using 2 min growth interruption at the GaAs well interfaces under arsenic

beam flux. The heterostructure was embedded in a pair of 72 nm $Al_{0.17}Ga_{0.83}As$ layers and further confined using $Al_{0.3}Ga_{0.7}As$ barriers. Experimental details of the PL measurements have been described previously [5].

3. Results and discussion

Figure 1(a)-(d) shows a cw PL spectrum taken at T = 4 K and an excitation wavelength of 697 nm as well as three PL excitation (PLE) spectra measured by detecting in the low-energy tails of the QW1, QW2, and QW3 emission bands. Sharply split excitonic emission and resonance absorption bands labeled A, B, C, and D are seen as a result of the formation of growth islands, whose lateral sizes are greater than the corresponding exciton Bohr radius [6]. We note that in the PL spectrum shown in Fig. 1(d) the split PL bands from QW2 and QW3 occurring at higher energies exhibit much higher intensities than the lowest energy peak of QW1, when the photoexcitation wavelength is close to the barrier band edge at 697 nm. This observation agrees with our previous study [2, 5] for a similar triple QW sample, but prepared without growth interruption, indicating the more efficient resonant capture of the photoexcited carriers by QW2 and QW3 rather than by QW1 under this excitation condition. The PL peaks are all red-shifted (by 1.4 ~ 2.4 meV) from the corresponding exciton resonances (as clearly observed in the PLE spectra) due to the intra-island exciton localization [7]. When comparing the three PLE spectra with each other, clear PLE correlations exist between the QW's [1]. In Fig. 1(a), a weaker PLE peak appearing around 728 nm is due to the absorption in the respective second (n=2) subband of QW1 [1]. This energy position is located below the band edge of the barrier, as clearly confirmed by a steep PL increase below 700 nm in the PLE spectra (b) and (c).

Figures 2(a) and 2(b) show PL time traces of the QW2 band and time-integrated PL spectra for

QW1 and QW2, respectively, measured at 15 K, when the photoexcitation wavelength of 782 nm is nearly resonant to the ground (n=1) excitonic state C of QW2. In these measurements, only PL signals cross-polarized to the linearly polarized laser light were recorded in order to suppress the scattered laser light coming into the streak camera. That is, when cold excitons with a negligible center-of-mass motion are resonantly photoexcited within the dominant growth island terrace C (cf. Fig. 1 for the island distribution), those excitons undergo energy relaxation towards the terraces B and A, loosing their energy by acoustic phonon emission and exciton-exciton scattering [8]. Thus, the split PL bands are observed for QW2 [Fig. 2 (b)]. However, the expected split PL lines are absent for QW1 under this condition. The energy relaxation in QW2 towards the wider-well terraces B and A is allowed at low temperatures, only when the excitons generated within the spatially localized area of the narrower-well terrace C can diffuse to other terraces, which are energetically lower than the initially occupied one. Delayed PL rise times of the exciton emissions B and A in Fig. 2(a) are thus explained by intra-well exciton localization towards the wider-well terraces [9], while the single-exponential decays (~0.4 ns) are attributed to the free exciton radiative recombination lifetimes [10].

Figure 3 shows TR-PL results, when the photoexcitation energy is tuned close to the n=2 state of QW1. In Fig. 3(a), the PL intensities of QW1 and QW2 are plotted on a logarithmic scale as a function of time, while in (b) time-integrated PL spectra are shown for QW1 (solid curve) and QW2 (dashed curve). We stress here that the PL intensity distribution between the QW's, the PL fine structure associated with the terraces, and the PL time-trace of each QW are all dramatically changed. For example, the PL fine structures due to the QW1 terraces appear with an enhancement of the PL intensity relative to the other wells. The contrast between Figs. 2 and 3 is remarkable. However, the fine structure of QW1 is smeared out for excitation energies outside the n=2 resonance. The PL fine structure observed for QW2 in Fig. 2(b) has disappeared in Fig. 3(b). In addition, the PL transients are also significantly modified,

indicating a non-exponential time dependence with prolonged rise and decay times. These interesting observations suggest that the exciton energy relaxation processes, particularly in QW1 are very different from the ones in the other two QW's.

When the QW1 layer is excited by the n=2 excitonic resonance absorption, more excitons in the excited state are generated within the QW1 layer with a negligibly small in-plane momentum ($K_{||} \sim 0$). Those excitons with small $K_{||}$ values must vertically relax to the ground states in momentum and real space. They are not efficiently trapped by the spatially localized potential minima or the trapping states because of the momentum conservation. Therefore, the PL intensity of the QW1 emission band is enhanced and the fine structure appears. Then, the prolonged rise time for QW1 observed in Fig. 3 suggests the vertical relaxation time constant longer than the radiative lifetime (τ_R) of the excitonic ground state, since in this case the rise time is basically determined by τ_R. We note that the transient PL intensity of the QW1 excitons reaches its maximum around t = 0.35 ns in Fig. 3 in general agreement with the experimentally observed values of τ_R. We also note that the efficient n=2 emission is observed in the PL spectra by employing high-energy excitation. The proposed mechanism based on the arguments of the center-of-mass motion of excitons is consistent with the disappearance of the PL splitting of QW1 outside the exciton resonance excitation, since under the off-resonant excitation conditions all excitons are generated with large $K_{||}$ values.

4. Conclusion

The photoluminescence properties of three electronically isolated GaAs QW's having different thicknesses with growth islands in each of them have been investigated by time-resolved photoluminescence experiments. It is found that the distribution of the PL intensity over the three

excitonic emission bands with monolayer splitting and their dynamics strongly depend on the energy state, in which the excitons are photogenerated, e.g. resonantly into the ground or first excited state. The origin of the different characteristics due to the exciton relaxation and radiative recombination processes has been discussed based on the center-of-mass motion of photoexcited excitons.

Acknowledgements

This work was supported in part by Grand-in-Aid for Scientific Research (No.11650020) from the Ministry of Education, Science, Sports, and Culture of Japan.

References

[1] L. Schrottke, H. T.Grahn, K. Fujiwara, Phys. Rev. B **56** (1997) 13321.

[2] K. Fujiwara et al., Inst. Phys. Conf. Ser. No 166 (IOP, Bristol, 2000) p. 103.

[3] A. Tomita, J. Shah, R.S. Knox, Phys. Rev. B **53** (1996) 10793.

[4] S.C. Hohng et al., Phys. Rev. B**60** (1999) 8883.

[5] K. Fujiwara, H.T. Grahn, L. Schrottke, K.H. Ploog, Proceedings of 25th International Conference on the Physics of Semiconductors (Osaka, Japan, 2000), to be published.

[6] U. Jahn et al., J. Cryst. Growth **150** (1995) 43.

[7] K. Fujiwara, H.T. Grahn, K.H. Ploog, Phys. Rev. B**56** (1997) 1081.

[8] R. Eccleston et al., Phys. Rev. B**45** (1992) 11403.

[9] K. Fujiwara et al., Phys. Rev. B **43** (1991) 13978.

[10] J. Feldmann et al., Phys. Rev. Lett. **59** (1987) 2337.

Figure captions

Fig. 1 PLE [solid curves in (a)-(c)] and PL [dashed curve in (d)] spectra of the triple QW sample at 4 K. The excitation wavelength for PL is 697 nm. The detection wavelengths for PLE are set at values of (a) 799 nm for QW1, (b) 788 nm for QW2 and (c) 771 nm for QW3. The spectra are shifted vertically for clarity.

Fig. 2 (a) Semi-logarithmic plots of the PL intensity as a function of time for the three excitonic emission bands, A, B, and C of QW2 measured at detection wavelength ranges of 785-787 nm, 783-785 nm, and 781-783 nm, respectively. A photoexcitation wavelength of 782 nm is used, which is resonant to the exciton absorption line C. (b) Time-integrated PL spectra normalized to the exciton peak B of the QW2 emission bands. The spectra are obtained by integrating the time period of 0.19-1.27 ns after the excitation pulse. The lattice temperature is 15 K and the averaged excitation power density of the 10 ps pulsed laser light is estimated to be a few W/cm^2.

Fig. 3 (a) Semi-logarithmic plots of the PL intensity as a function of time for the excitonic emission bands of QW1 and QW2 measured at detection wavelength ranges of 792-801 nm and 781-790 nm, respectively. A photoexcitation wavelength of 735 nm, which is nearly resonant to the first excited state of QW1, is used. (b) Time-integrated PL spectra, normalized to the exciton peak B of the QW1 for QW1 and QW2. The lattice temperature is 15 K and the averaged excitation power density of the 10 ps pulses laser light is estimated to be a few W/cm^2.

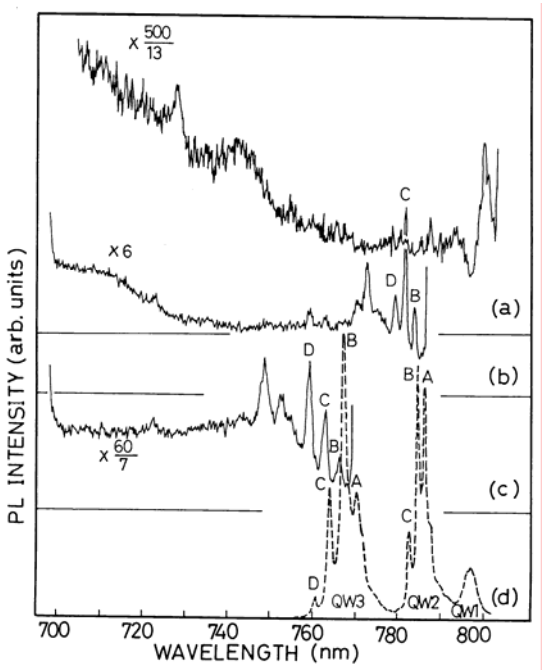

Fig. 1

K. Fujiwara et al., Physica E (ARW)

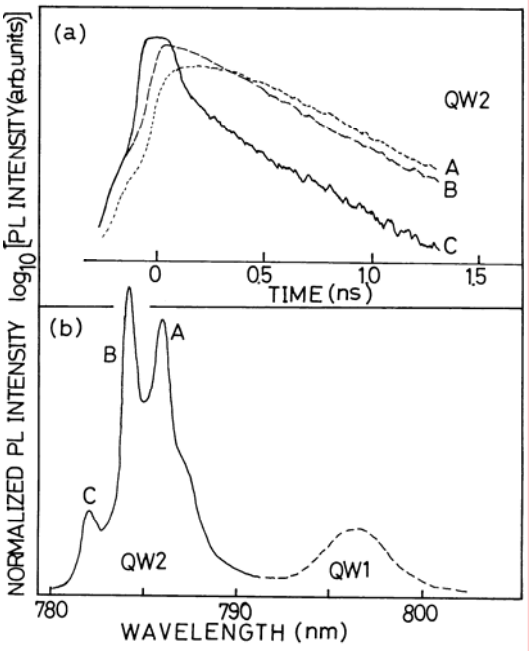

Fig. 2

K. Fujiwara et al., Physica E (ARW)

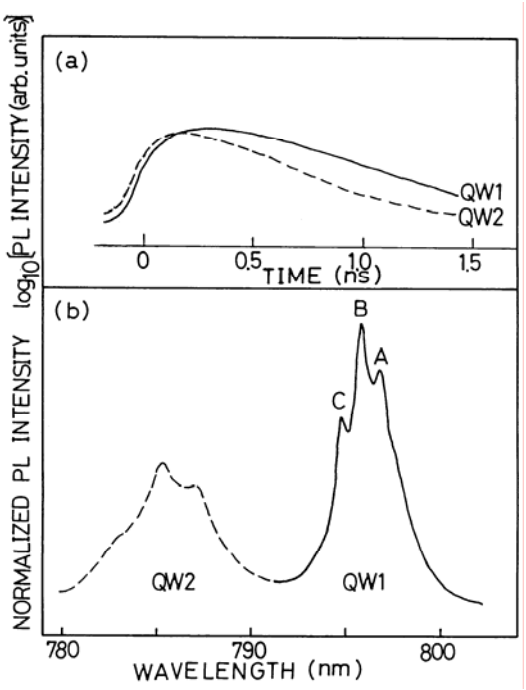

Fig. 3

K. Fujiwara et al., Physica E (ARW)