PHYSICAL REVIEW B VOLUME 42, NUMBER 15 15 NOVEMBER 1990-II

Well-width dependence of photoluminescence excitation spectra in GaAs-$Al_xGa_{1-x}As$ quantum wells

N. Ogasawara,* A. Fujiwara, and N. Ohgushi
Department of Applied Physics, Faculty of Engineering, The University of Tokyo, Bunkyo-ku, Tokyo, Japan

S. Fukatsu, Y. Shiraki, Y. Katayama,† and R. Ito
Research Center for Advanced Science and Technology, The University of Tokyo, Meguro-ku, Tokyo, Japan

(Received 12 July 1990)

Photoluminescence excitation spectra of high-quality GaAs-$Al_{0.3}Ga_{0.7}As$ quantum wells with various widths have been measured at 77 K. It has been found that the luminescent intensity for excitation photon energies exceeding the barrier band-gap energy exhibits an oscillatory behavior against the well width. The results clearly show that the trapping efficiency of the excited electrons into a well strongly depends on the well width through the resonance and off resonance of the quantum-well levels with the conduction-band bottom of the barrier.

Recent advances in growth technology of semiconductor quantum wells (QW's) by molecular-beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) have stimulated the research on physical phenomena arising from the size quantization of electronic systems. Among them, it is of great significance to investigate the process of electron transfer from the bulk states into the quantized states in the well since the process plays important roles in such novel devices utilizing the size quantization as quantum-well lasers and resonant tunneling diodes.

A notable example of these investigations is the observation of the anomalous excitation intensity dependence of photoluminescence in GaAs-$Al_xGa_{1-x}As$ QW's.[1,2] There, the exponent m in the excitation intensity (I_{ex}) dependence of the luminescent intensity (I_l), $I_l \propto I_{ex}^m$, was shown to vary ($m=1-2$) periodically with well width L_z. The phenomenon was interpreted in terms of quantum oscillation of electron trapping efficiency into a well; under resonant conditions (i.e., when the band bottom of the barrier energetically degenerates to one of the quantum levels) which appear periodically with L_z, the trapping efficiency was assumed to be reduced since carriers easily escape from the well, and then, using a rate equation of excited carriers, the periodic variation of m was analyzed to be correlated to the variation of the trapping efficiency.[1-3] However, the interpretation is not intuitive since the exponent m is determined as a result of competition among various relaxation processes.[1-3]

In this paper we report on the first measurement of photoluminescence excitation spectra (PLES) of QW's with excitation photon energies covering both above and below the barrier band-gap energy and show that the PLES measurement of QW's with various widths is a useful tool in gaining more direct insight into the well-width dependence of the carrier trapping efficiency. It is demonstrated that the luminescent intensity exhibits an abrupt increase as the excitation photon energy exceeds the barrier band-gap energy. The increase occurs since a large amount of carriers excited in the barrier region are relaxed down to the wells and contribute to the luminescence in the well. Therefore the amount of the increase in the luminescent intensity gives direct information on the carrier trapping efficiency. The observation of this phenomenon in QW's with various well widths reveals that the carrier trapping efficiency indeed depends strongly on the well width through the resonance and off-resonance of the quantum-well levels with the conduction-band bottom of the barrier.

The sample measured was MBE-grown GaAs-$Al_{0.3}Ga_{0.7}As$ crystal comprising six GaAs wells with a series of widths L_z, 17–155 Å, separated from each other by 500-Å-thick $Al_{0.3}Ga_{0.7}As$ barriers. As reported previously,[1,2] MBE growth of GaAs-$Al_xGa_{1-x}As$ QW's at relatively low substrate temperatures (~580 °C) using a zone-refined high-purity Al source provides QW wafers with extremely smooth heterointerfaces featuring narrow photoluminescence line widths. The smoothness of heterointerfaces is obviously essential in investigating size-quantization phenomena.[1,2] The sample used here was cleaved out from the same wafer as those used in the previous studies.[1,2]

The PLES were taken at 77 K with excitation by a DCM (4 dicyanomethylene-2-methyl-6-*p*-dimethylaminostyryl-4H-pyran) dye laser pumped by a multiline Ar-ion laser. Photoluminescence with a fixed excitation photon energy consisted of six peaks (1.52–1.73 eV) corresponding to the electron-heavy hole transitions with the lowest quantum number ($n=1$) of the six wells. The intensity of each peak was measured as a function of the dye laser frequency to obtain PLES. The dye laser power incident on the QW sample was adjusted by a neutral density filter and was kept constant at 5 mW when the excitation photon energy was varied in the range of 1.75–2.05 eV. The excited area was estimated to be 0.015 mm^2.

Figures 1(a)–1(d) show the PLES of the QW's with the well width $L_z = 31$ Å (a), 52 Å (b), 77 Å (c), and 104 Å (d), respectively, where E_b (~ 1.85 eV) denotes the band-gap energy of the barrier. It can be seen in all the figures that the luminescent intensity I_l exhibits an abrupt increase at the excitation photon energy $\hbar\omega_{\mathrm{ex}} = E_b$. This is understood as follows. For $\hbar\omega_{\mathrm{ex}} < E_b$, photogeneration of carriers occurs only within the thin wells since the barriers are transparent. In contrast, as $\hbar\omega_{\mathrm{ex}}$ exceeds E_b, a large number of carriers is generated in the barrier region because the barriers (500 Å) are much thicker than the wells. A significant part of the excited carriers are relaxed into the wells and contribute to the luminescence, resulting in a remarkable increase in I_l. It should be noted in Fig. 1 that the amount of the increase varies significantly with L_z. This indicates that the efficiency of carrier trapping into a well depends strongly on L_z.

In order to gain further insight into the L_z dependence of the trapping efficiency, I_l for $\hbar\omega_{\mathrm{ex}}$ just above E_b (I_{lb}) and below E_b (I_{lw}) are plotted as a function of L_z in Fig. 2(a). Here, the experimental I_{lb} value of each well (Fig. 1) was multiplied by a factor appropriate to the well to compensate for the excitation-light extinction in the barriers and to make it thereby possible to compare I_{lb} for a common effective I_{ex} among the wells. The excitation-light absorption coefficient in the barrier was assumed to

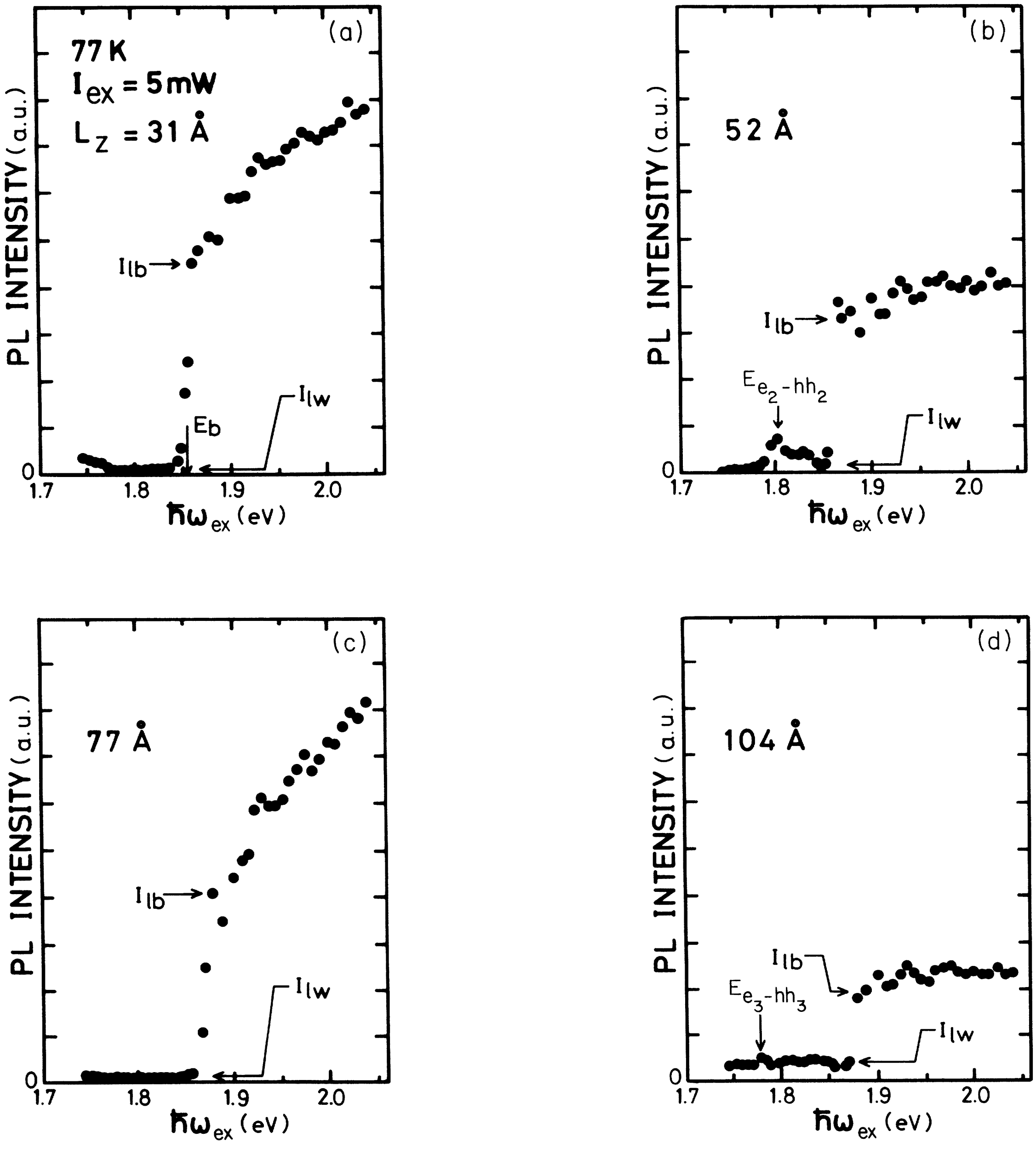

FIG. 1. Photoluminescence excitation spectra of MBE-grown GaAs-Al$_{0.3}$Ga$_{0.7}$As quantum wells at 77 K with the excitation power of 5 mW. The well widths are (a) 31 Å, (b) 52 Å, (c) 77 Å, and (d) 104 Å, respectively. E_b: the band-gap energy of the barrier. $E_{e_2\text{-hh}_2}$, $E_{e_3\text{-hh}_3}$: the transition energy between electron and heavy hole levels with $n = 2, 3$.

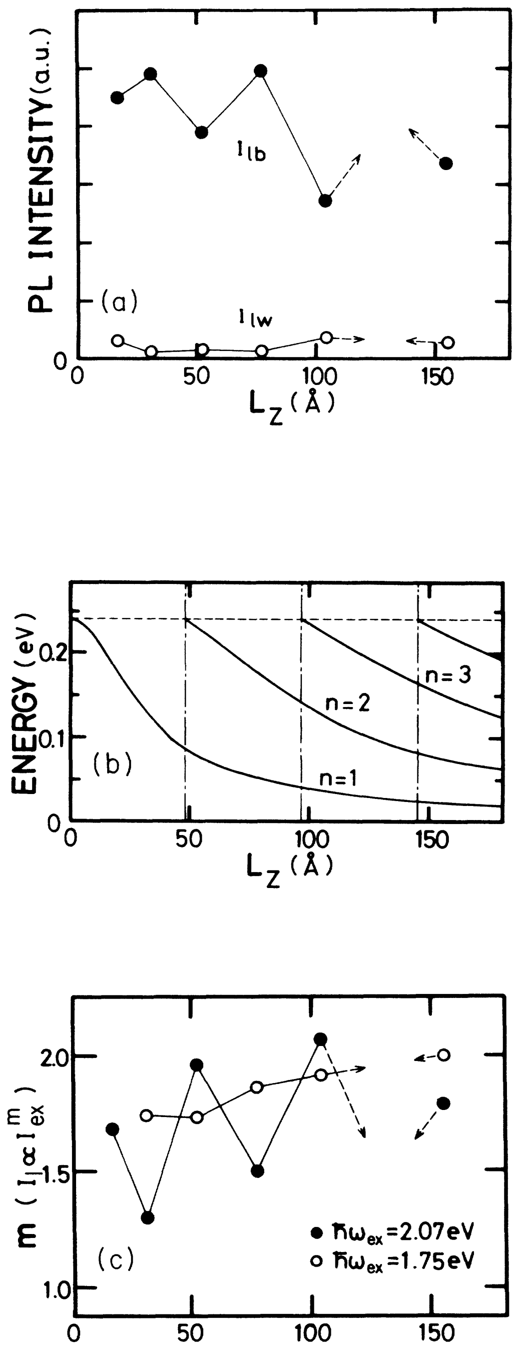

FIG. 2. (a) Luminescent intensities for excitation photon energies $\hbar\omega_{ex}$ just above the band-gap energy of the barrier E_b (I_{lb}: the solid circles) and below E_b (I_{lw}: the open circles) vs well width L_z. Both the solid and dashed lines are to guide the eye. (b) Variations of quantized energy levels of electrons (quantum number n) with L_z (the solid lines). The conduction-band offset ΔE_c is depicted by the dashed line. The dash-dotted lines depict the resonant conditions. (c) The exponent m of the excitation intensity (I_{ex}) dependence of luminescent intensity (I_l), $I_l \propto I_{ex}^m$, for $\hbar\omega_{ex}=2.07$ eV ($>E_b$) (the solid circles) and $\hbar\omega_{ex}=1.75$ eV ($<E_b$) (the open circles) as a function of L_z. m was determined from the measurements in the excitation power range of 0.1–10 mW. Both the solid and dashed lines are to guide the eye.

be 1×10^4 cm^{-1} (Ref. 4) and the correction factor, which is a monotonically increasing function of L_z since the wells are located in the order of increasing L_z from the light-incident top surface, was 1.42 ($L_z=155$ Å) at maximum. It can be seen in Fig. 2(a) that I_{lb} exhibits a periodic variation against L_z with a large amplitude in contrast to I_{lw}, which shows no systematic variation. The oscillatory behavior of I_{lb} evidently represents an oscillation of carrier trapping efficiency against L_z. It is to be mentioned here that the periodic variation of I_{lb} was reproducibly observed in several samples with essentially the same structure showing that the variation is indeed intrinsic. In contrast, the small variation of I_{lw} depended on samples and can be presumably attributed to the variation of the crystal quality among the wells.

Figure 2(b) helps us in viewing the oscillation of I_{lb} in relation to the size quantization, where the calculated L_z dependences of the quantized energy levels of electrons (quantum number n) are shown by the solid lines. The conduction-band discontinuity ΔE_c (the dashed line) was taken to be 65% (0.24 eV) of the band-gap discontinuity ΔE_g (0.37 eV), consistent with recent studies.[5–7] The electron effective mass was taken to be $0.067m_0$.[8] Comparing Fig. 2(a) with 2(b), one can see that I_{lb} is minimized (i.e., the trapping efficiency is minimized) under the resonant conditions (the dash-dotted lines) where the highest quantized level coincides with the bottom of the conduction band. This is in agreement with the simple picture;[1,2] under the resonant conditions, electrons which approach the well can readily reescape from or pass through the well because of the continuity of the energy level while, under off-resonant conditions, they are tightly confined and contribute to the luminescence after the relaxation down to the $n=1$ subband.

The solid circles and the open circles in Fig. 2(c) depict the exponent m that represents I_{ex} dependence of I_l ($I_l \propto I_{ex}^m$) for $\hbar\omega_{ex}=2.07$ eV ($>E_b$) and $\hbar\omega_{ex}=1.75$ eV ($<E_b$), respectively. It can be seen that, for $\hbar\omega_{ex}=2.07$ eV, m shows a periodic variation essentially identical to that observed in the previous experiments[1,2] using an Ar-ion laser as the excitation-light source [$\hbar\omega_{ex}=2.4$–2.7 eV ($>E_b$)]. The coincidence of the period between m ($\hbar\omega_{ex}=2.07$ eV) and I_{lb} supports the conclusion[1,2] that the oscillation of m ($\hbar\omega_{ex}>E_b$) originates from the well-width dependence of electron trapping efficiency. The absence of periodic variation in m ($\hbar\omega_{ex}=1.75$ eV) offers a further support since the trapping process is not involved in the luminescence with $\hbar\omega_{ex}<E_b$.

It is worth mentioning here that whether the trapping efficiency is reduced[3] or enhanced[9] under the resonant conditions has been a subject of theoretical controversy. The discrepancy among the theoretical studies seems to stem from the difficulty in describing theoretically the electron transfer from the three-dimensional states in the barrier into the two-dimensional states in the well. The present paper, however, has provided clear experimental evidence of trapping-efficiency reduction under the resonant conditions. In the theoretical study by Murayama[3] only the quantization of electrons was taken into account and a three-dimensional band approximation was employed for holes. The approximation seems to be qualitatively plausible because the energy separation among hole quantized levels is smaller. In fact, the experimental results presented in the previous[1,2] and present papers can be interpreted within the framework of the approximation. However, further theoretical and experimental studies are certainly needed to quantitatively elucidate the effect of hole quantization.

It is interesting to note that I_l shows virtually no dispersion for $\hbar\omega_{\mathrm{ex}} > E_b$ under the resonant conditions [Figs. 1(b) and 1(d)] while, under the off-resonant conditions [Figs. 1(a) and 1(c)], I_l gradually grows as $\hbar\omega_{\mathrm{ex}}$ is increased further beyond E_b. At present, there exists no reasonable explanation for the difference. This behavior is one of the problems that should be studied both experimentally and theoretically to deepen our understanding of the electron transfer in QW's.

In conclusion, PLES of QW's have been measured over the excitation photon energy range covering both above and below the barrier band-gap energy for the first time and the well-width dependence of carrier trapping efficiency has been firmly confirmed. Further details including the temperature dependence of PLES will be reported elsewhere.

The authors wish to thank Dr. T. Mishima of Central Research Laboratory, Hitachi Ltd. for supplying the QW samples.

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[*] Present address: Department of Electronics Engineering, University of Electro-Communications, 1-5-1 Chofugaoka, Chofu, Tokyo 182, Japan.

[†] Also at Optoelectronics Technology Research Laboratory, 5-5 Tohkodai, Tsukuba, Ibaraki 300-26, Japan.

[1] T. Mishima, J. Kasai, M. Morioka, Y. Sawada, Y. Murayama, Y. Katayama, and Y. Shiraki, in *12th International Symposium on GaAs and Related Compounds, Karuizawa, 1985*, Inst. Phys. Conf. Ser. No. 79, edited by M. Fujimoto (Hilger, London, 1986), Chap. 8, p. 445.

[2] T. Mishima, J. Kasai, M. Morioka, Y. Sawada, Y. Katayama, Y. Shiraki, and Y. Murayama, Surf. Sci. **174**, 307 (1986).

[3] Y. Murayama, Phys. Rev. B **34**, 2500 (1986).

[4] M. D. Sturge, Phys. Rev. **127**, 768 (1962).

[5] H. Kroemer, W. Y. Chien, J. S. Harris, Jr., and D. D. Edwall, Appl. Phys. Lett. **36**, 295 (1980).

[6] R. C. Miller, A. C. Gossard, D. A. Kleinman, and O. Munteanu, Phys. Rev. B **29**, 3740 (1984).

[7] D. Arnold, A. Ketterson, T. Henderson, J. Klem, and H. Morkoç, J. Appl. Phys. **57**, 2880 (1985).

[8] Q. H. F. Vrehen, J. Phys. Chem. Solids **29**, 129 (1968).

[9] G. Bastard, Phys. Rev. B **30**, 3547 (1984).