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Photoluminescence of an $Al_{0.5}Ga_{0.5}As/GaAs$ multiple quantum well in the temperature range from 5 to 400 K

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Photoluminescence (PL) of an unintentionally doped $Al_{0.5}Ga_{0.5}As/GaAs$ multiple quantum well (MQW) has been measured at temperatures from 5 to 400 K. It was found that the ratio of the intensity of the $n=1$ electron-light hole transition ($1e$-$1lh$) to that of the $n=1$ electron-heavy hole transition ($1e$-$1hh$) can be described by an exponential function of reciprocal temperature. The excitation-power dependence of the $1e$-$1hh$ transition PL intensity measured at temperatures from 5 to 296 K in steps of 15–20 K showed that the relative contribution of free-carrier recombination gradually increases from 5 to 120 K and then remains constant. This tendency was confirmed by the temperature dependence of the energy difference between the $1e$-$1hh$ transition and the bulk GaAs band gap. © *2009 American Institute of Physics*. [doi:10.1063/1.3256222]

I. INTRODUCTION

Beginning with the pioneering work by Dingle *et al.*,[1] a large number of studies have been devoted to the optical properties of AlGaAs/GaAs quantum wells (QWs). Photoluminescence (PL) at different temperatures,[2–5] PL excitation (PLE) spectroscopy,[6] and absorption[7] in these structures have all been intensively studied. As a result, AlGaAs/GaAs QWs found widespread applications in optoelectronic devices.[8]

In the present work, we measured the PL spectra of an $Al_{0.5}Ga_{0.5}As/GaAs$ multiple QW (MQW) over a wide temperature range from 5 to 400 K. In the spectra reported in this study and in Refs. 2–4, the strongest luminescence is caused by the transition from the $n=1$ electron sublevel to the $n=1$ heavy hole sublevel; this transition is denoted as $1e$-$1hh$. The ratio of the intensity of the $n=1$ electron-light hole transition ($1e$-$1lh$) to that of the $1e$-$1hh$ transition is known to increase with temperature due to the thermal population of holes from the $1hh$ sublevel to the $1lh$ sublevel.[2] To our knowledge, no model has yet been put forward to describe this phenomenon quantitatively. This study proposes a model in which the temperature dependence of the above ratio can be described by an exponential function of reciprocal temperature. The model was verified against experimental observations in the temperature range 40–340 K.

The mechanism of PL at room temperature in AlGaAs/GaAs QWs has been a topic of extensive debate. Most authors[2,9,10] attributed the dominant PL mechanism at room temperature to excitonic recombination. However, Fouquet and Siegman[11] and Zhongying *et al.*[4] asserted that free-carrier recombination should dominate the PL spectra. On the other hand, Hayakawa *et al.*[5] concluded that the PL is the result of both free-carrier and excitonic recombination. A common approach to determining the recombination mechanism is to observe the dependence of the PL intensity I_{PL} on the excitation power I_{ext}. This dependence can be described by the formula[4,11,12] $I_{PL}=C\times I_{ext}^{t}$, where C is a constant, and the exponent t has a value of 1 for excitonic recombination and a value of 2 for free-carrier recombination.[4,11] In previous studies,[4,5,11] such excitation-power dependence measurements have been performed at only a few temperatures, which did not allow a detailed investigation of the variation of the PL mechanism. In this study, the excitation-power dependence was measured from 5 to 296 K in steps of 15–20 K. As a result, changes in the PL mechanism could be clearly observed in the temperature dependence of the t exponent. Observations revealed that, at low temperature, excitonic recombination was the dominant PL mechanism of the $1e$-$1hh$ transition. As the temperature was increased in the range from 5 to 120 K, the contribution of free-carrier recombination to the total PL intensity was found to increase. At higher temperatures in the range from 120 to 296 K, the relative contribution remained constant. The temperature dependence of the energy difference between the $1e$-$1hh$ transition and the bulk GaAs band gap was similar to that of the t exponent. Both dependences support the claim by Hayakawa *et al.*[5] that both excitonic and free-carrier recombination are responsible for PL emission in AlGaAs/GaAs MQWs at room temperature.

II. EXPERIMENT

The sample used in this study was grown on a (100)-oriented GaAs substrate by molecular beam epitaxy. It consisted of a 0.5-μm-thick $Al_{0.5}Ga_{0.5}As$ buffer layer, 100 periods of alternating $Al_{0.5}Ga_{0.5}As$ and GaAs layers and a 6-nm-thick GaAs cap layer. The grown layers were unintentionally doped. The $Al_{0.5}Ga_{0.5}As$ barrier width was close to 10 nm and the GaAs well width, determined from the PL peaks (see below), was 9.6 ± 0.6 nm (17 ± 1 monolayers). The sample was cooled in a vacuum cryostat by means of a closed system utilizing liquid He. The temperature of the sample was varied from 5 to 400 K.

In the PL measurements, the conventional configuration was used. The sample was excited with a 635 nm line of a laser diode. The PL emission was focused onto the slit of a 0.3-m-diffraction-grating monochromator, and a 100

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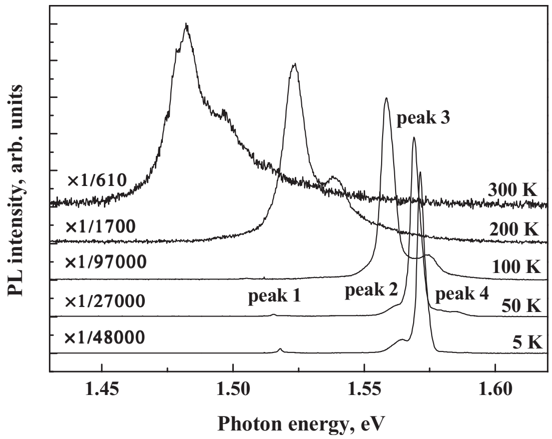

FIG. 1. PL spectra of the $Al_{0.5}Ga_{0.5}As/GaAs$ MQW at different temperatures.

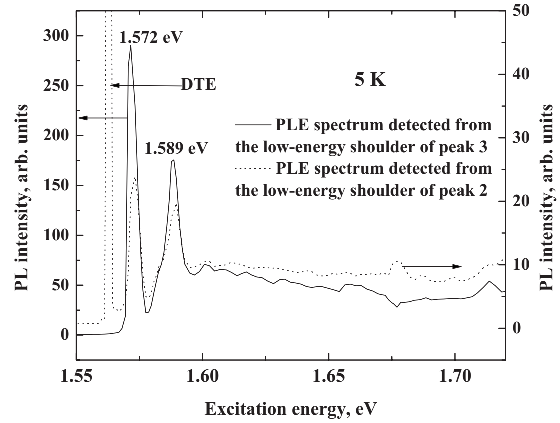

FIG. 3. PLE spectra detected from the low-energy shoulder of peaks 2 and 3 at 5 K.

×1340 pixel charge-coupled device (CCD) array was used to detect the PL signal. In the excitation-power dependence measurements, output power of the laser diode was varied with an attenuator and was registered by means of an optical power meter. In the PLE measurements, the excitation source was a Ti:sapphire laser, whose wavelength was varied in the range 716–820 nm with a step of about 1 nm. PL intensity was normalized by excitation power.

III. RESULTS AND DISCUSSION

A. Photoluminescence spectra and peak assignment

The PL spectra from the MQW are shown in Fig. 1. At 5 K, three peaks are observed at 1.518, 1.565, and 1.572 eV. At this temperature, the 1.572 eV peak has the highest intensity. As the temperature is increased, an additional peak appears on the high-energy side of the strongest peak. At 50 K, four separate peaks can be distinguished; they are denoted as peak 1, peak 2, peak 3, and peak 4 in order of increasing energy, as shown in Fig. 1. The energy difference between peak 4 and peak 3 at 50 K is 15 meV.

In Fig. 2, the energy of each peak is plotted as a function of temperature and compared to the bulk GaAs band gap energy.[13] It can be seen that the curves for peaks 1, 3, and 4

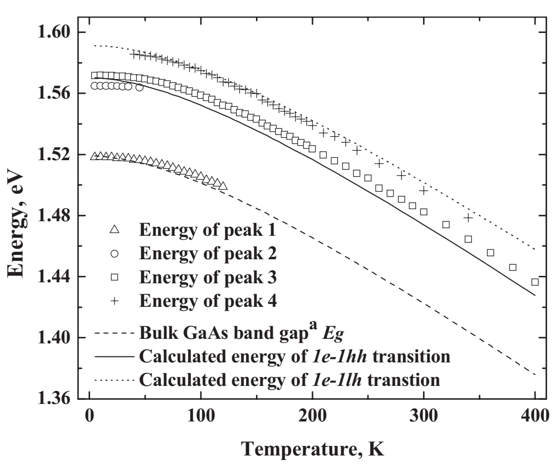

FIG. 2. Energies of peaks 1–4, the bulk GaAs band gap (Ref. 13), and the calculated 1*e*-1*hh*, 1*e*-1*lh* transitions vs temperature.

follow the general shape of the bulk GaAs band gap energy curve, shown by the dashed line. The curve for peak 1 is very close to the bulk GaAs band gap energy curve and can therefore be attributed to PL from the bulk GaAs substrate. In addition, peaks 3 and 4 can be attributed to the 1*e*-1*hh* and 1*e*-1*lh* transitions,[2,3] respectively. Figure 2 also shows the calculated energies for the 1*e*-1*hh* (solid line) and 1*e*-1*lh* (dotted line) transitions in the $Al_{0.5}Ga_{0.5}As/GaAs$ MQW. The calculations were carried out based on the Kronig–Penney model using the values of the band offsets, light hole masses, and electron masses given in Ref. 14 and the heavy hole masses given in Ref. 15. According to the calculations, a well width of 9.6 ± 0.6 nm (17 ± 1 monolayer) gives the best fit to the experimental data.

Figure 3 illustrates PLE spectra detected from the low-energy shoulder of peaks 2 and 3, hereafter referred to as PLE spectra from peaks 2 and 3, respectively. The PLE spectrum from peak 3 shows two distinguishable features at 1.572 and 1.589 eV, which can be ascribed to the 1*e*-1*hh* and 1*e*-1*lh* free excitons[6] (FEs), respectively. In the PLE spectrum from peak 2, the two FE features are observed at the same energy positions. Additionally, the PL intensity from peak 2 drastically decreases at excitation energies just below the 1*e*-1*hh* FE energy (but still above the detection energy). Regarding these excitation energies, exciton formation can occur only through low-probability phonon-assisted mechanisms. As a result, the exciton production rate is expected to decrease, leading to a considerable reduction in the PL intensity of excitons bound to impurities. Therefore, peak 2 could be attributed to recombination of impurity-bound excitons.

Thermal quenching of the integrated PL intensity of the 1*e*-1*hh* transition is depicted in Fig. 4. The experimental data in the temperature range 5–200 K were fitted using the expression[2,4] $I_{PL}=A/[1+B\exp(-E_a/kT)]$, to obtain the activation energy $E_a=14.1\pm0.7$ meV.

B. Temperature dependence of the ratio of the PL peak intensity of the 1*e*-1*lh* transition to that of the 1*e*-1*hh* transition

It is presumed that the PL peak intensity I is proportional to the hole concentration n_h. Assuming a Boltzmann distribution, $n_h \propto \exp[-(F_v-E_h)/kT]$, where F_v is the nonequilib-

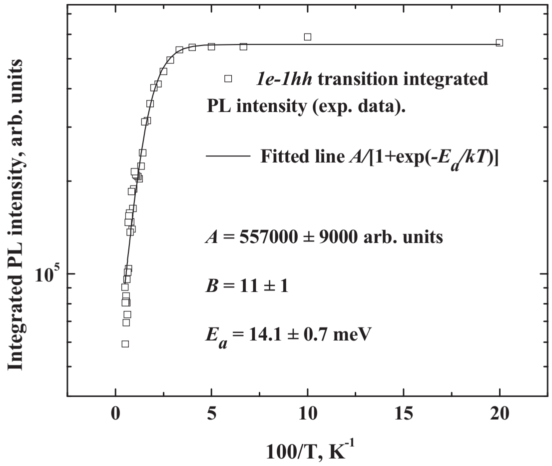

FIG. 4. Temperature dependence of the integrated PL intensity of the $1e$-$1hh$ transition.

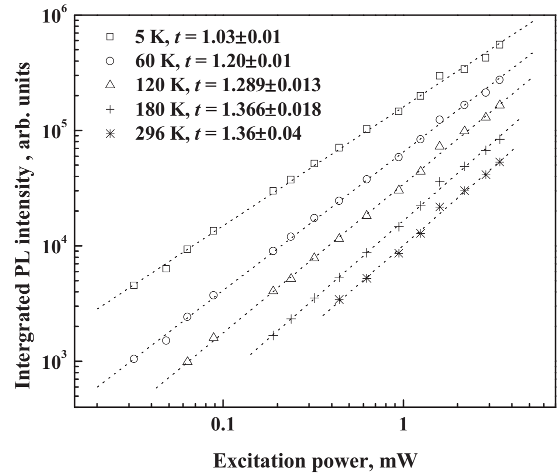

FIG. 6. Dependence of integrated PL intensity of peak 3 on excitation power at 5, 60, 120, 180, and 296 K. The t exponent in the expression $I_{PL}=CI_{ext}^t$ is also shown.

rium quasi-Fermi level in the valence band and E_h is the hole energy level. As a result, the PL peak intensities of the $1e$-$1hh$ and $1e$-$1lh$ transitions are given as $I_{hh}=\alpha_{hh}(T)\exp(-(F_v-E_{1hh})/kT)$ and $I_{lh}=\alpha_{lh}(T)\exp(-(F_v-E_{1lh})/kT)$, where E_{1hh}, E_{1lh} are the energies of the $1hh$ and $1lh$ sublevels, respectively, and α_{hh}, α_{lh} are coefficients. Assuming

$$\alpha_{lh}(T)/\alpha_{hh}(T) = 1 \tag{1}$$

and taking into account the fact that the difference in energy between the $1hh$ and $1lh$ sublevels is equal to that between the $1e$-$1lh$ and $1e$-$1hh$ transitions, the ratio I_{lh}/I_{hh} can be written as

$$I_{lh}/I_{hh} = \exp(-\Delta E/kT), \tag{2}$$

where $\Delta E=E_4$-E_3 and E_3, E_4 are the energies of peaks 3 (the $1e$-$1hh$ transition) and 4 (the $1e$-$1lh$ transition), respectively.

Figure 5 shows the ratio I_{lh}/I_{hh} plotted versus reciprocal temperature with a semilogarithmic scale. The data are fitted by Eq. (2) with ΔE calculated from the experimental values for the energies of peaks 3 and 4. The theoretical curve is a close fit to the experimental data except for the data points at the lowest (40 K) and highest (340 K) temperatures. At 40 K,

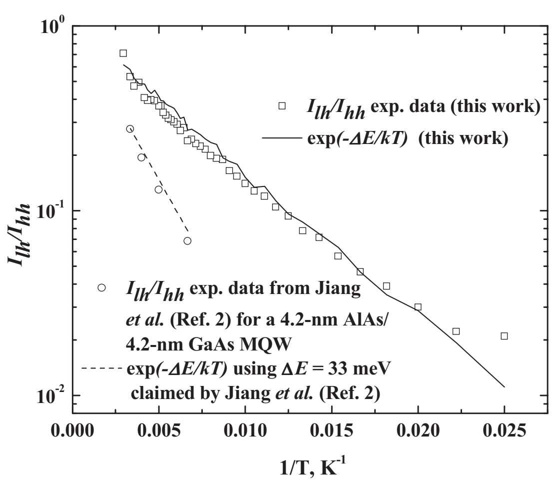

FIG. 5. Temperature dependence of the ratio I_{lh}/I_{hh}.

it is possible that the PL of peak 4 at 1.586 eV was affected by a known defect in the CCD array at 1.590 ± 0.002 eV. At 340 K, the accuracy of the measured PL intensities for peaks 3 and 4 is expected to be poor because of the low signal-to-noise ratio. In general, the data are most reliable in the temperature range 70–260 K, where peaks 3 and 4 are clearly resolved. Figure 5 also shows experimental I_{lh}/I_{hh} data derived from the PL spectra of a 4.2 nm AlAs/4.2 nm GaAs MQW reported by Jiang *et al.*[2] Again, these data are approximated by Eq. (2) using the 33 meV energy difference between the $1e$-$1lh$ and $1e$-$1hh$ transitions claimed in the reference. The approximation line fits the experimental data well, which suggests that Eqs. (1) and (2) are valid regardless of the barrier aluminum mole factor and the well width.

In the case of free-carrier recombination, the assumption that the PL peak intensity is proportional to the hole concentration is reasonable, since PL intensity is proportional to the product of the electron and hole concentrations.[16] For excitonic recombination, this cannot be so readily assumed, because in this case the PL intensity is proportional to the exciton concentration.[16] Since an exciton is an electron-hole pair, the exciton concentration should be proportional to either the electron or hole concentration. To fulfill the charge neutrality condition, the $n=1$ electron concentration should be larger than both the $n=1$ heavy hole concentration and the $n=1$ light hole concentration. Therefore, the exciton concentration should again be proportional to the hole concentration as required. In addition, as the temperature increases, peaks 3 and 4 partly overlap due to thermal broadening. Even though this fact was not considered in the model, Eq. (2) has proven to be a good approximation for the ratio I_{lh}/I_{hh} at temperatures in the range 50–300 K.

C. Dependence of integrated PL intensity on excitation power

The dependence of the integrated PL intensity on excitation power was measured in the temperature range 5–296 K in steps of 15–20 K. In Fig. 6, the integrated PL intensity of peak 3 at temperatures of 5, 60, 120, 180, and 296 K is

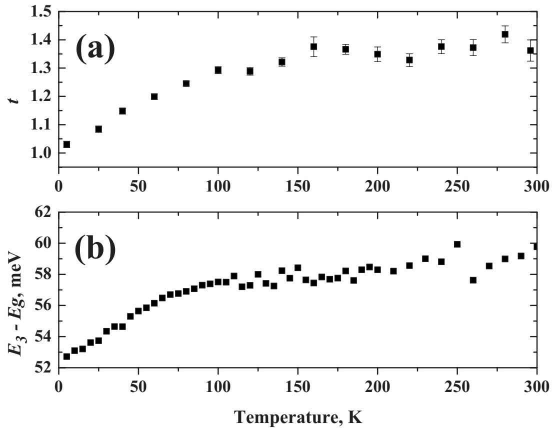

FIG. 7. Temperature dependence of (a) the t exponent and (b) energy difference between peak 3 and the bulk GaAs band gap.

plotted versus excitation power with a double-logarithm scale. The experimental data are fitted by straight lines, whose slopes are equal to the exponent t in the expression $I_{PL}=CI_{ext}^{t}$. At 5 K, $t=1.03$, indicating that the dominant recombination mechanism is excitonic at this temperature. Figure 7(a) illustrates the temperature dependence of the t exponent. As the temperature increases from 5 to 120 ± 20 K, the value of t increases from 1.03 to 1.37 ± 0.5. At temperatures from 120 ± 20 to 300 K, t remains constant at 1.37 ± 0.5. The deviation of the data point at 120 K ($t=1.29$) from the above trend may have been related to the accuracy of the measurement. The temperature dependence of t suggests that as the temperature increases from 5 to 120 ± 20 K, excitons are dissociated and free-carrier recombination gradually makes a larger contribution to the total PL intensity of the $1e$-$1hh$ transition. Above 120 ± 20 K, the relative contributions from free-carrier recombination and, thus, from excitonic recombination to the total PL intensity do not vary. This appears to validate Hayakawa's conclusions[5] on the presence of two PL recombination mechanisms at room temperature.

A large variance exists in the values of the t exponent reported in the literature. Fouquet and Siegman[11] found that $t=2$ at room temperature. The values of 1.05 at 11 K and 1.42 at 120 K reported by Zhongying *et al.*[4] are very close to the values 1.03 at 5 K and 1.38 at 160 K found in this study. However, Zhongying *et al.*[4] reported a further increase in t up to 1.91 at 200 K, in contrast with the results described here. In addition, Hayakawa *et al.*[5] reported a t value of 1.21 at 300 K for a 50 Å wide $Al_{0.3}Ga_{0.7}As/GaAs$ MQW. The discrepancy in the published results suggests that the t exponent is sensitive to the measurement conditions. Also, in this study, such sensitivity may have led to unreliability in some of the data points for the t exponent, for example, the one at 120 K. However, this is unlikely to have affected the overall trend observed in Fig. 7(a), due to the large experimental data set in the current study.

Let us denote the difference in energy between peak i ($i=1,3,4$) and the bulk GaAs band gap as E_i-E_g. Figure 7(b) plots the energy difference E_3-E_g in the temperature range 5–300 K. The shape of the curve is very similar to that of the temperature dependence of the t exponent except for the fact that the former stabilizes at 105 ± 10 K and the latter at 120 ± 20 K. If a single recombination mechanism dominated the PL in the entire 5–300 K temperature range, the energy difference would not be expected to change in that temperature range. The fact that two completely different physical quantities show similar temperature dependence strongly supports the conclusions on the variation of the PL mechanism and on the relative contribution of free-carrier recombination to the total PL intensity. The relative contribution is expected to increase with temperature in the entire 5–300 K temperature range, since excitons are more easily dissociated at higher temperature. However, the observed stabilization of the t exponent and of the energy difference between peak 3 and the bulk GaAs band gap completely contradicts this expectation.

It should be noted that the temperatures 120 ± 20 and 105 ± 10 K correspond to thermal energies (kT) of 10.4 ± 1.7 and 9.0 ± 0.9 meV, respectively. These energies are close to the reported binding energies of the $1e$-$1hh$ exciton[6,8,17,18] in AlGaAs/GaAs MQWs with well widths almost identical to that used in this study. As a result, one can speculate that the temperature at which the t exponent and the energy difference E_3-E_g begin to stabilize may represent the binding energy of the $1e$-$1hh$ exciton. However, this leads to difficulties in explaining the fact (data not shown here) that the energy differences E_1-E_g and E_4-E_g also increase from 5 to 105 K before becoming constant, as in the case of E_3-E_g. Such similar stabilization temperatures would not be expected since the binding energies of bulk GaAs, $1e$-$1hh$, and $1e$-$1lh$ excitons are different. For example, the binding energy of bulk GaAs is claimed to be 4 meV,[1] whereas the binding energies of $1e$-$1hh$ and $1e$-$1lh$ excitons in an $Al_{0.4}Ga_{0.6}As/GaAs$ QW with a width of 9.6 nm were calculated to be 10 and 13 meV,[18] respectively. More investigations are needed to clarify this problem.

IV. CONCLUSION

The PL properties of an $Al_{0.5}Ga_{0.5}As/GaAs$ MQW were investigated at temperatures in the range 5–400 K. The ratio of the intensity of the PL peak due to the $1e$-$1lh$ transition to that due to the $1e$-$1hh$ transition could be described qualitatively by the formula $\exp(-\Delta E/kT)$. This was verified by comparison with experimental data in the temperature range 50–300 K.

In order to determine the PL recombination mechanism, the dependence of the PL intensity on excitation power was measured over a wide temperature range from 5 to 296 K in small steps of 15–20 K. From the results, it was concluded that the relative contribution of free-carrier recombination to the total $1e$-$1hh$ PL intensity increased with temperature in the range 5–120 K, and then remained constant from 120 to 296 K. It is hoped that this work will be of benefit in the design of AlGaAs/GaAs MQW-based optoelectronic devices.

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