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Temperature Dependence of the Photoluminescence in GaAs-GaAlAs Multiple Quantum Well Structures

By

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The temperature dependence of the photoluminescence (PL) of GaAs/GaAlAs multiple quantum wells is investigated. Emissions related to transitions between $n = 1, 2, 3$ electron and hole subbands are observed. Theoretical evaluation of the energy levels fits nicely the experimental data. The temperature dependence of peak position and photoluminescence intensity and the dependence of PL intensity on the power excitation show that there are three different temperature regions. At low temperatures (22 to 40 K) the main emission is ascribed to exciton recombination while at temperatures higher than 100 K it can be attributed to free carrier recombination. In the intermediate temperature region the PL involves both, excitonic and free carrier recombination. Thermally activated non-radiative recombination processes strongly reduce the luminescence quantum efficiency.

Es wird die Temperaturabhängigkeit der Photolumineszenz (PL) von GaAs/GaAlAs-Mehrfachquantenwells untersucht. Emissionen, die mit Übergängen zwischen $n = 1, 2, 3$ Elektronen- und Löcher-Subbänder verknüpft sind, werden beobachtet. Eine theoretische Entwicklung der Energieniveaus stimmt gut mit den experimentellen Werten überein. Die Temperaturabhängigkeit der Lage des Maximums und Photolumineszenzintensität und die Abhängigkeit der Pl-Intensität von der Anregung zeigt, daß drei verschiedene Temperaturbereiche existieren. Bei niedrigen Temperaturen (22 bis 40 K) wird die Hauptemission einer Exzitonenrekombination zugeschrieben, während bei Temperaturen höher als 100 K sie der Rekombination freier Träger zugeordnet wird. Im mittleren Temperaturbereich schließt die PL sowohl Exzitonen- als auch freie Ladungsträger-Rekombination ein. Ein thermisch aktivierter, nichtstrahlender Rekombinationsprozeß reduziert die Lumineszenzquantenausbeute.

1. Introduction

During the past few years it has been possible to grow systems consisting of alternate layers of two different semiconductors with controlled thickness and relatively sharp interfaces using epitaxial crystal growth techniques such as molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD). A great deal of efforts has been devoted to the study of these new one-dimensional periodic structures. Recently many papers [1 to 5] have been published discussing PL emission; in fact the study of the luminescence properties of quantum well (QW) structures provides

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basic knowledge for the manufacturing of QW photo-electronic devices such as QW lasers, QW photo-detectors and QW photo-conductive switches.

Low temperature investigation of PL has shown that, for high quality quantum well samples, it can be mainly attributed to excitonic recombinations [6]. On the other hand, significant differences in the PL spectra have been reported for room temperature measurements. Some authors [7] claimed that, even at room temperature, the main PL spectra are due to radiative recombination of free excitons while other authors stated [8] that room temperature luminescence is due to free carrier radiative recombinations.

In this paper we report a detailed study of the temperature dependence of the photoluminescence in GaAs–GaAlAs multiple quantum well structures. At low temperature we observed the transitions of intrinsic excitons related to electron subbands (E_{1e}, E_{2e}, and E_{3e}) and hole subbands (E_{1hh}, E_{1lh}, E_{2hh}, and E_{3hh}). The experimental results fit nicely to the theoretical calculations of the subband energy levels. The theoretical calculations were made assuming a finite potential well depth and nonparabolic GaAs Γ_6^c conduction band [9].

Measurements at different temperatures of the energy peak positions, power excitation dependence of the PL intensity, and temperature dependence of the PL intensity show that there are three temperature regions. The first region can be related to the intrinsic exciton recombinations (low temperatures), the second to both intrinsic excitonic and free carrier recombinations, and the third one to free carrier recombinations (high temperatures); the three regions have different thermally activated non-radiative recombination mechanisms.

Another interesting aspect of the luminescence is that the intensity ratio between the second subband and the first one decreases as the temperature increases. This behaviour is anomalous with respect to usual photoluminescence spectra of multiple quantum well structures. We believe that this is due to strong non-radiative recombinations at high temperatures present in this sample.

2. Sample and Experimental Apparatus

The sample studied in this work was grown by molecular beam epitaxy (MBE) technique. The sample consists of the following layer sequence. A GaAs buffer layer was grown first on the Cr-doped semi-insulating GaAs(100) substrate, followed by a 0.1 μm thick GaAlAs layer and 12 periods of alternating 12 nm GaAs well layers and 40 nm $Ga_{1-x}Al_xAs$ barrier layers with $x = 0.4$.

Conventional PL techniques are used in the experiment. The sample held in a closed cycle helium cryostat (Air products DE 202S) was excited by an argon ion laser (Spectra Physics model 165) operating at 514.5 nm with a maximum power of ≈ 4 mW. The area of the focused beam spot was about 0.5 mm^2. The luminescence was analyzed by means of a Spex double monocromator (model 1680) and detected by a cooled Hamamatsu RS 943-02 photomultiplier with a GaAs cathode. The signal was measured by means of a picoammeter, converted from analog to digital and sent to an Apple II E computer for elaboration.

3. Results and Discussion

3.1 Photoluminescence spectra

Fig. 1 shows a typical low temperature (22 K) PL spectrum. The three peaks at about 1.540, 1.611, and 1.755 eV, denoted as E_{1h}, E_{2h}, and E_{3h} are attributed to the expected $n = 1$, $n = 2$, and $n = 3$ electron–heavy hole intrinsic excitonic recombina-

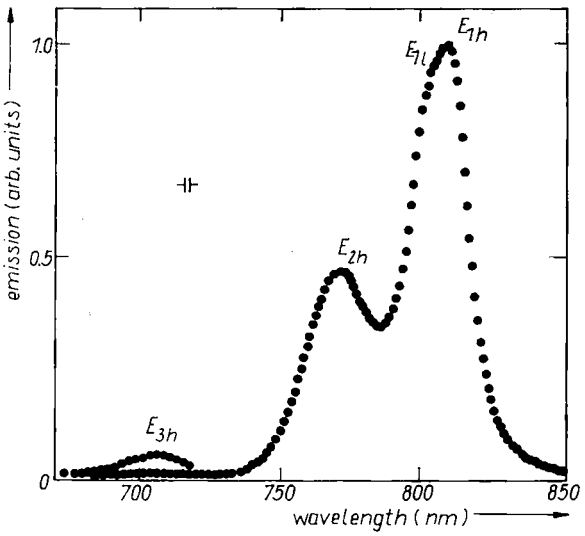

Fig. 1. Low temperature ($T = 22$ K) PL spectrum of GaAs/GaAlAs MQW; $P = 4$ mW

tions. The shoulder at ≈ 1.552 eV of the first peak is due to the E_{1l} electron–light hole intrinsic excitonic transition.

In order to assign the observed peaks we calculated the energy position of the subbands in a quantum well structure according to quantum theory. As it is well known, when the well thickness L_z compares with the de Broglie wavelength of the carriers, the energy levels will be quantized in the z direction. For a finite potential depth V_0 the Schrödinger equation has a solution for E_n values given by

$$\beta_n = \alpha_n \tan \alpha_n \qquad (n = 1, 3, 5 \ldots)\,, \tag{1a}$$

$$-\beta_n = \alpha_n \cot \alpha_n \qquad (n = 2, 4, 6 \ldots)\,, \tag{1b}$$

where

$$\alpha_n = \left(\frac{L_z^2 m_1 E_n}{2\hbar^2}\right)^{1/2}, \tag{1c}$$

$$\beta_n = \left[\frac{L_z^2 m_2 (V - E_n)}{2\hbar^2}\right]^{1/2}, \tag{1d}$$

and m_1 is the effective mass of carriers in GaAs and m_2 is the effective mass of carriers in GaAlAs.

The well depth for the electrons and holes has been taken [10] as

$$V_c = 0.85\, \Delta E_g, \qquad V_v = 0.15\, \Delta E_g\,,$$

where

$$\Delta E_e = E_{g(\mathrm{GaAlAs})} - E_{g(\mathrm{GaAs})}\,.$$

In the energy level calculation we take into account the non-parabolicity of the GaAs Γ_6^c conduction band assuming for the electron effective mass the following energy dependence [10]:

$$m_e(E) = (0.0665 + 0.0436E + 0.236E^2 - 0.147E^3)\, m_0\,, \tag{1e}$$

where m_0 is the free electron mass. Using this formula the energy of higher order subbands is reduced. In the calculation we have also considered the exciton binding

Table 1
The calculated and experimental value of the subband levels (meV)
$L_z = 12$ nm $T = 22$ K

n	E_{ne}	E_{nhh}	E_{nh}*)	E_{nh}(exp)
1	28	5	25	22
2	103	18	113	93
3	196	39	227	237

*) $E_{nh} = E_{ne} + E_{nhh} - E_{ex}$.

energy [6] as a function of the well depth and L_z. Theoretical and experimental values are summarized in Table 1.

Fig. 2 a and b show the temperature dependence of the PL spectra. When increasing the temperature, we observe an increase of the intensity ratio between the E_{1h} and E_{2h} bands. This behaviour is opposite to what normally occurs in the PL of MQW's.

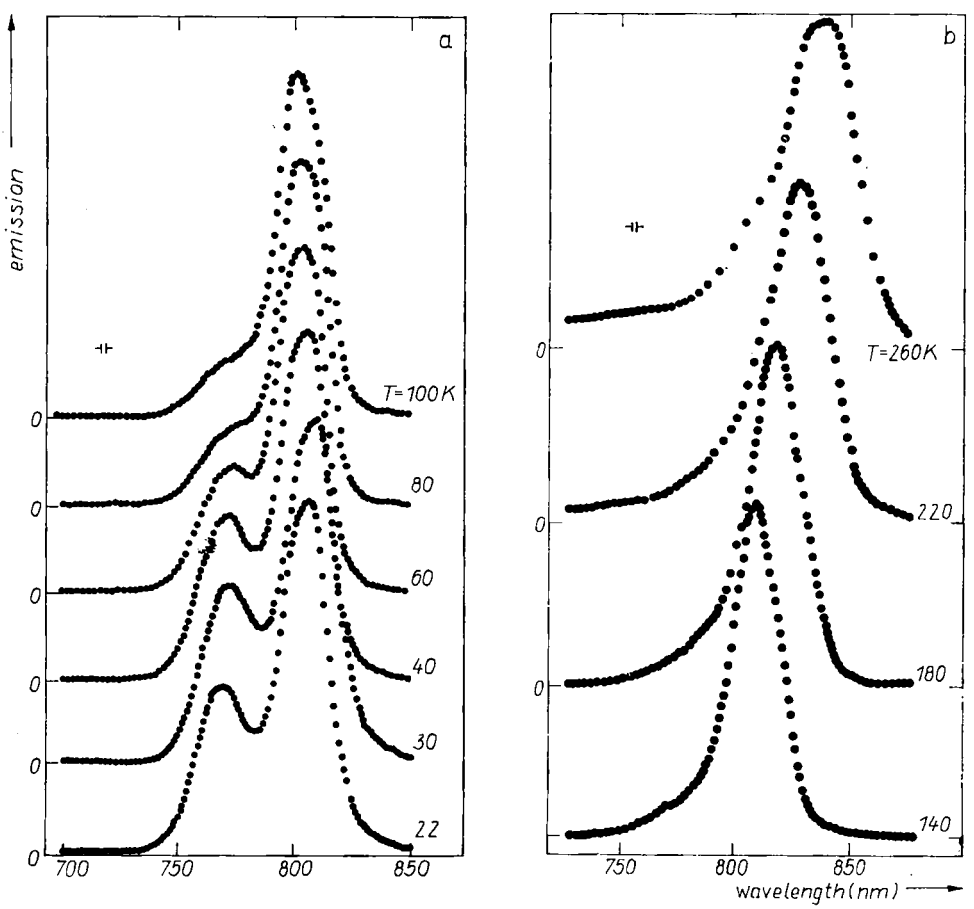

Fig. 2. Photoluminescence spectra at different temperatures

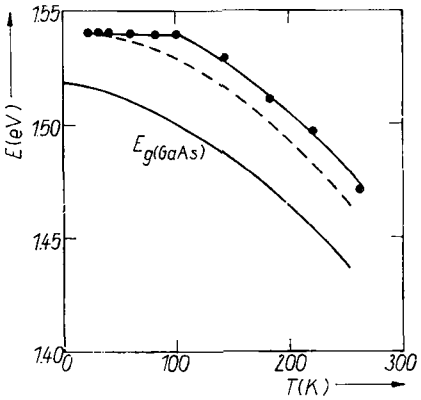

Fig. 3. Temperature dependence of the E_{1h} energy peak position (dots are experimental data). Also shown are the temperature dependences of the GaAs energy gap and of $E_{g(GaAs)} + E_{1e} + E_{1hh} - E_{ex}$ (dashed curve)

We believe that this feature arises from a strong non-radiative recombination present at high temperatures and we will attempt a plausible explanation later on.

3.2 Temperature dependence of the position of the E_{1h} band

Fig. 3 shows the measured temperature dependence of the E_{1h} energy peak position. We have drawn also in the same figure the temperature dependence of the band gap of bulk GaAs calculated according to the expression

$$E_{g(GaAs)} = 1.519 - \frac{5.405 \times 10^{-4} T^2}{(204 + T)} \text{ (eV)} . \tag{2a}$$

Three temperature regions can be inferred from Fig. 3. From 22 to 40 K the experimental data fit the dashed line

$$E_{1h} = (E_{1e} + E_{1hh}) + E_{g(GaAs)} - E_{ex} \tag{2b}$$

which confirms that the main emission is due to exciton recombination. (The values of E_{1e}, E_{1hh} are given in Table 1; E_{ex} is the exciton binding energy.) From 40 to 100 K the peak energy of the E_{1h} band satisfies the condition

$$(E_{1e} + E_{1hh}) + E_{g(GaAs)} - E_{ex} < E_{1h} < (E_{1e} + E_{1hh}) + E_{g(AGaAs)} \tag{2c}$$

and therefore the emission is due to both excitonic and band-to-band recombinations. From 100 K to room temperature the E_{1h} band peak can be fitted by the expression

$$E_{1h} = (E_{1e} + E_{1hh}) + E_{g(GaAs)} \tag{2d}$$

which suggests that the main emission is due to free carrier recombinations from the conduction subbands to the valence subbands.

3.3 Dependence of the PL intensity on the excitation power

Fig. 4 shows the dependence of the PL intensity on the excitation power at three different temperatures 25, 100, and 203 K. The excitation power was varied from 0.09 to 4 mW.

The excitation power P dependence of the PL intensity I turns out to be a power law: $I \sim P^{\alpha}$. The exponent factors α were found to be 1.09, 1.58, and 1.98 at 25, 100, and 203 K, respectively. This behaviour can be easily explained if one considers the rate equations for the recombination processes.

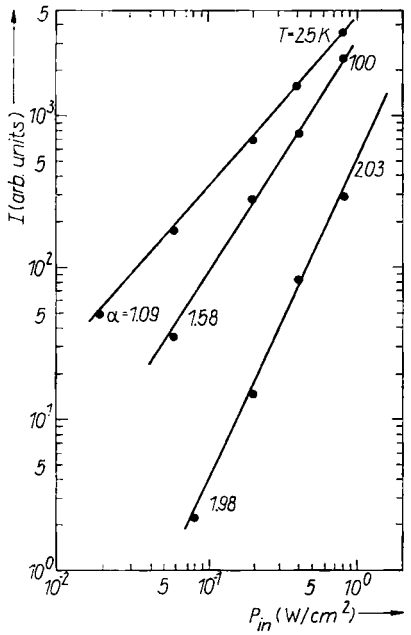

Fig. 4. Excitation dependence of the integrated photoluminescence intensity at different temperatures. The exponent α in equation $I_{\mathrm{pl}} = CP_{\mathrm{in}}^{\alpha}$ is also indicated

In fact free exciton recombinations must follow a first-order kinetic [8]

$$\frac{\mathrm{d}N_{\mathrm{x}}}{\mathrm{d}t} = G - (\gamma_{\mathrm{rx}} + \gamma_{\mathrm{nrx}})\, N_{\mathrm{x}} , \tag{3a}$$

where N_{x} is the exciton population, G the pump rate, and γ_{rx} and γ_{nrx} are the radiative and non-radiative recombination rates, respectively. The photon emission rate is given by

$$I(t) = \gamma_{\mathrm{rx}} N_{\mathrm{x}}(t) . \tag{3b}$$

Solving (3a) for steady-state condition, we obtain

$$N_{\mathrm{x}} = G\tau_{\mathrm{x}} , \tag{3c}$$

where τ_{x} is the exciton lifetime and $G \sim P$ so that we have $I \sim G \sim P$. The observed dependence $I \sim P^{1.09}$ at 25 K confirms that at low temperature the emission is due to free exciton recombination.

At higher temperatures the carrier recombination kinetic is no longer first-order so the rate equation becomes

$$\frac{\mathrm{d}n}{\mathrm{d}t} = G - Bnp - \gamma_{\mathrm{nre}} n , \tag{3d}$$

where n and p are the free electron and hole concentrations, Bp is the radiative recombination rate and γ_{nre} the non radiative recombination rate for electrons. Assuming a small intrinsic carrier density for our undoped sample, when non-radiative decay dominates, ($\gamma_{\mathrm{nre}} \gg Bp$), and considering the steady-state condition, expression (3d) becomes

$$G = \gamma_{\mathrm{nre}} n . \tag{3e}$$

The emission intensity for free carriers will be

$$I = Bnp \tag{3f}$$

so that $I \sim np \sim P^2$. The exponent factor $\alpha = 2$ observed at 203 K confirms that the photoluminescence is due to free carrier recombination associated with strong non-radiative recombinations.

In the intermediate temperature region we can assume that the emission involves both mechanisms.

3.4 Temperature dependence of PL intensity

Fig. 5 shows the temperature dependence of the PL intensity. At low temperature the PL intensity is almost constant, while it drops very fast approaching room temperature.

The temperature dependence of the non-radiative transition rate can be written

$$\frac{1}{\tau_{nr}} = \frac{1}{\tau_{n\infty}} \exp\left(\frac{-E_v}{kT}\right), \tag{4a}$$

where E_v is the thermal activation energy. If the radiative recombination rate is written as

$$R_r = \frac{1}{\tau_r} \tag{4b}$$

the internal efficiency of luminescence η will become

$$\eta = \frac{1/\tau_r}{1/\tau_r + 1/\tau_{n\infty} \exp(-E_v/kT)} \tag{4c}$$

and the PL intensity can be written as

$$I = \frac{I_0}{1 + \tau_r/\tau_{n\infty} \exp(-E_v/kT)}. \tag{4d}$$

One gets from (4d)

$$\ln\left(\frac{I_0}{I} - 1\right) = -\frac{E_v}{kT} + \ln\frac{\tau_r}{\tau_{n\infty}}. \tag{4e}$$

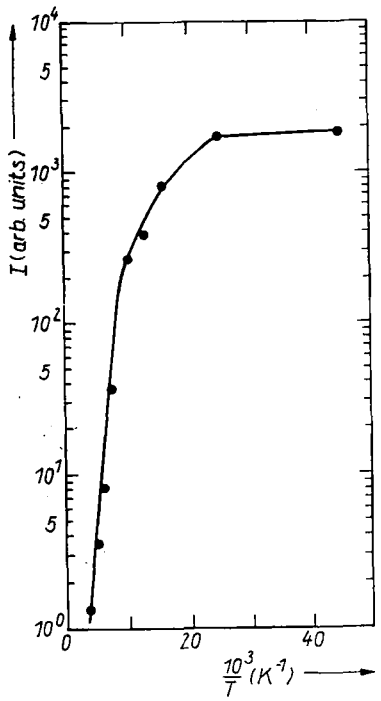

Fig. 5. Temperature dependence of the luminescence intensity

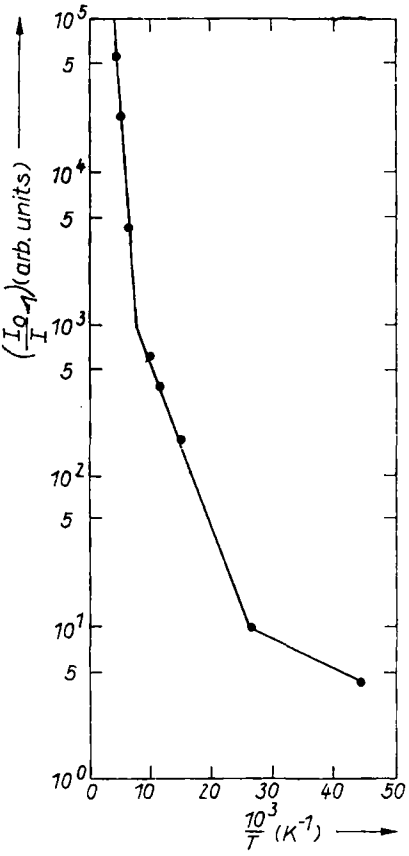

Fig. 6. Temperature dependence of ln $(I_0/I - 1)$. The slope of the curve gives the activation energies E_v (see the text)

$(I_0/I) - 1$ versus $1/T$ is plotted in Fig. 6 on a semilogarithmic scale. We can evaluate three E_v values from Fig. 6; between 22 and 40 K we obtain $E_v = 7$ meV, which is just the free exciton binding energy. This means that the main non-radiative process in this temperature region is the exciton dissociation process. From 40 to 100 K we obtain $E_v = 21$ meV and from 100 K to room temperature $E_v = 79$ meV. In the last two regions there are therefore two thermally activated non-radiative processes. At high temperatures the non-radiative recombination is very strong, the carrier population in the subbands decreases and the emission from the second subband disappears.

3.5 Comparison with other results

A number of papers has been recently published dealing with photoluminescence of GaAs–GaAlAs multiple quantum well structures. On the other hand, most attention has been devoted to extrinsic transitions originated from defect states, impurities, interface states, and traps [11 to 14].

To our knowledge a detailed analysis of the intrinsic PL of MQW structures under cw excitation as far as the temperature dependence of the peak positions and photoluminescence intensity and the dependence of the PL intensity on the excitation power is concerned has been only reported in [5]. The results we obtained show a nice overall agreement with those reported in [5].

4. Conclusions

We studied the temperature dependence of the PL from a MQW structure. At low temperature we observed the transition of intrinsic excitons related with electron subbands E_{1e}, E_{2e}, and E_{3e} and hole subbands E_{1hh}, E_{1lh}, E_{2hh}, and E_{3hh}. The experimental results are in good agreement with the theoretical calculations. At higher temperatures the radiative recombination of free excitons and free carriers contributes to the luminescence; finally at room temperature is mostly due to the radiative recombination of free carriers.

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