Thermal escape of carriers out of GaAs/$Al_xGa_{1-x}As$ quantum-well structures

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(Received 24 February 1992)

Nonradiative recombination processes in thin GaAs/$Al_xGa_{1-x}As$ quantum-well (QW) and double-barrier quantum-well structures have been investigated by means of continuous-wave and time-resolved photoluminescence (PL) measurements. We find that, due to the temperature dependence of the radiative time constant, the Arrhenius plot of the PL intensity cannot be used to extracting the activation energy of the nonradiative channels. In fact it is the temperature dependence of the PL decay time T_L which directly gives information on the thermal activation of the loss mechanism. The activation energies obtained from the Arrhenius plot of T_L demonstrate that, at least in the case of thin wells, the main nonradiative mechanism is the thermal escape of the less-confined species of carriers.

I. INTRODUCTION

Semiconductor quantum wells (QW) are known to act as efficient traps for the carriers photogenerated by optical absorption. The electron-hole pairs created in the barrier region rapidly relax their excess of energy, through the interaction with phonons, and are trapped into the confined states of the wells, from where they eventually recombine either radiatively or not radiatively. The investigation of the carrier trapping processes in QW's has received increasing attention in the last few years, and several papers have been devoted to carrier trapping from both the theoretical[1,2] and experimental[3,4] points of view. It is well known, in addition, that the collection efficiency determines, to a large extent, the performances of quantum-well photonic devices and that it can be optimized by growing additional confinement layers,[3] such as in nonconventional structures like separated confinement quantum wells, widely used for reducing the threshold current of QW lasers.[5]

Less attention has been devoted to the inverse mechanism, i.e., detrapping or escape of carriers out of the QW's, even if a reduction of the carrier collection efficiency in the high-temperature[4,6] and high excitation[4,7] regimes has been reported. Only recently the photoluminescence (PL) quenching of $In_xGa_{1-x}As$/GaAs QW's has been interpreted[8,9] in terms of the thermal emission of electron-hole pairs from the quantum wells into the barriers, on the basis of a fair agreement between the activation energy, extracted from the Arrhenius plot of the PL intensity, and the excitonic confinement energy. Nevertheless some striking points, such as the effect of the temperature dependence of the radiative recombination rate or the escape of free carriers, rather than excitons, have not yet been discussed.

In this paper we present a systematic study of the nonradiative recombination in a set of thin GaAs/$Al_xGa_{1-x}As$ quantum wells and double-barrier quantum wells (DBQW). The analysis of the temperature dependence of both the PL decay time T_L and the integrated-PL intensity I_L allows us to study the nonradiative time constant T_{NR}. We show that, due to the strong temperature dependence of the radiative time constant T_R, the Arrhenius plot of I_L cannot be used for finding the activation energy of the nonradiative channels. Indeed it is the temperature dependence of T_L that contains the information about the nonradiative recombination time T_{NR}. From the Arrhenius plot of T_L we find activation energies much smaller than the electron-hole-pair confinement energies but in good agreement with the unipolar escape out of the wells of the less confined species of carriers. This result, on one hand, shows that thermal escape is, at least in the case of thin GaAs/$Al_xGa_{1-x}As$ wells, the main nonradiative process and, on the other hand, it demonstrates that the free-carrier escape is more efficient than that of excitons. Finally the comparison of the thermal activation energies with the confinement energies results in a better agreement if a band offset ratio $\Delta E_c:\Delta E_v = 70:30$ is used.

II. SAMPLES AND EXPERIMENT

We have investigated two different sets of samples. The first consists in two pairs of GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells of widths 20 and 40 Å, respectively. The second set consists in GaAs/AlAs/$Al_{0.3}Ga_{0.7}As$ double-barrier quantum wells with widths of 20 and 40 Å including either 1 or 2 AlAs monolayers (ML) embracing the wells. Details of the growth procedures have been described elsewhere.[10,11] In the following we will refer to the first two samples as 0 ML-1 and 0 ML-2, respectively, while the two DBQW structures will be denoted as 1 ML and 2 ML.

We have used a synchronously pumped mode-locked dye laser for the time-resolved measurements; the time duration of the pulses was 3 ps, with a repetition rate of 76 MHz and the wavelength tuned to $\lambda_{exc}=5700$ Å. The luminescence was analyzed with two different detection techniques. A streak camera system having a time resolution of the order of 20 ps and a time window of 1.5 ns has been used in the case of decay times T_L shorter than 500 ps, while we have used a time-correlated single-photon counting apparatus for higher values of T_L, providing a time resolution of the order of 100 ps. Continuous-wave photoluminescence and photoluminescence excitation measurements (PLE) have been performed at 4 K using an Ar^+ pumped dye laser (620–670 nm). The PL signal was dispersed by a double monochromator and analyzed by usual photon counting detection. The samples were held in a variable temperature cryostat (4–300 K) and the excitation intensity was always kept around 10 W/cm^2 in order to avoid screening and band filling effects.

Typical PL spectra at low temperature ($T=4$ K) of the QW's investigated are reported, as solid lines, in Fig. 1. The most evident feature of these spectra is the increase of the excitonic recombination energy by adding the AlAs monolayers. In the case of the thinnest wells (20 Å), we find a shift of the PL lines of more than 100 meV from the 0 ML to the 2 ML DBQW. This shift directly reflects the increase of the carrier confinement energy due to the insertion of the ultrathin AlAs barriers; indeed, DBQW's are allowed to have very shallow subbands even in relatively thick wells.[10,11] For instance, the carrier confinement energy of the 20-Å DBQW with 2 AlAs monolayers is of the same order as that of a 8-Å single quantum well. This peculiar feature of DBQW's makes such structures ideal systems for studying the carrier dynamics in the limit of very shallow subbands. We also remark that the PL-integrated intensity of the 20-Å DBQW's is of the same order of magnitude as that of the other QW's investigated, while a reduction of the PL efficiency has been reported for thin SQW's of $In_xGa_{1-x}As/GaAs$,[8,9] having subbands of similar shallowness as in our case.

In Fig. 1 we also report, as dotted lines, the PLE spectra of the 40-Å QW's with photon energies around the $Al_{0.3}Ga_{0.7}As$ absorption edge. From the energy position of the excitonic peaks of the alloy we are able to obtain

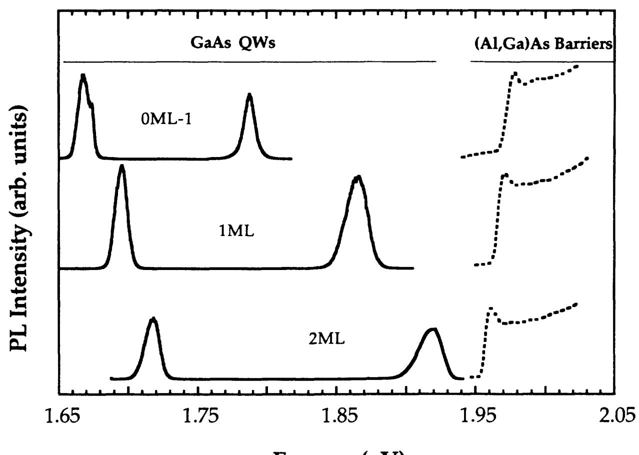

FIG. 1. PL spectra (solid lines) and PLE spectra of 40-Å wells (dotted lines) for the 0 ML-1, 1 ML, and 2 ML samples, respectively. Sample 0 ML-2 shows a similar behavior as 0 ML-1.

the band discontinuity ΔE_g of each sample investigated. The electron-hole-pair confinement energy $\Delta_{e\text{-}h}$, reported in Table I, has then been estimated by subtracting from ΔE_g the energy of the PL peaks from the QW's after taking into account the calculated exciton binding energy.[10]

The temperature dependence of the PL decay time of the QW's investigated is reported in Fig. 2. It should be noted that, at low temperature, the thinnest DBQW's (20 Å; 1–2 ML's) show a value of T_L larger by a factor 2 with respect to the other wells investigated. We interpret this feature as a signature of the decrease of the excitonic oscillator strength: in fact, as discussed in Ref. 11, the insertion of the AlAs monolayers produces an increase of the confinement energy and, consequently, a delocalization of the excitonic wave function. At the same time, the larger value of T_L is an indication of the fact that the nonradiative mechanisms play a negligible role in the carrier recombination at low temperature.

On the other hand, the general features of the temperature dependence of the PL decay times reported in Fig. 2 are very similar to those previously reported in the literature.[9,12,13] Raising the temperature T_L starts increasing and, after reaching a maximum value, which strongly de-

TABLE I. Parameters of the QW's investigated. The activation energies E_A are extracted from the fits of Fig. 5; the electron-hole-pair confinement energies $\Delta_{e\text{-}h}$ are measured by comparing the PL spectra and the PLE spectra reported in Fig. 1; the electron (hole) confinement energies Δ_e (Δ_h) are calculated by fitting $\Delta_{e\text{-}h}$ with an effective-mass model using two different band offset ratios: (a) $\Delta E_c:\Delta E_v=70:30$; (b) $\Delta E_c:\Delta E_v=65:35$.

Sample	L_W (Å)	$\Delta_{e\text{-}h}$ (meV)	E_A (meV)	Δ_h (meV) (a)	Δ_e (meV) (a)	Δ_h (meV) (b)	Δ_e (meV) (b)
2 ML	20	45	20	31	14	43	3
1 ML	20	105	50	49	56	60	46
0 ML-1	20	190	70	69	123	87	103
0 ML-2	20	200	65	71	128	90	107
2 ML	40	240	100	90	150	110	130
1 ML	40	270	90	97	172	116	173
0 ML-1	40	300	90	103	197	124	180
0 ML-2	40	300	90	103	197	124	180

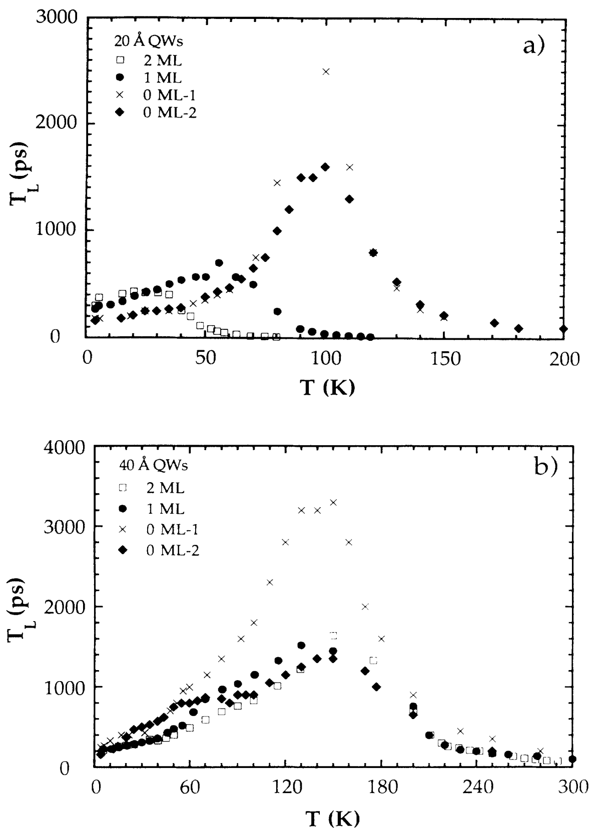

FIG. 2. Temperature dependence of the PL decay time T_L of the wells investigated: (a) 20-Å QW's; (b) 40-Å QW's.

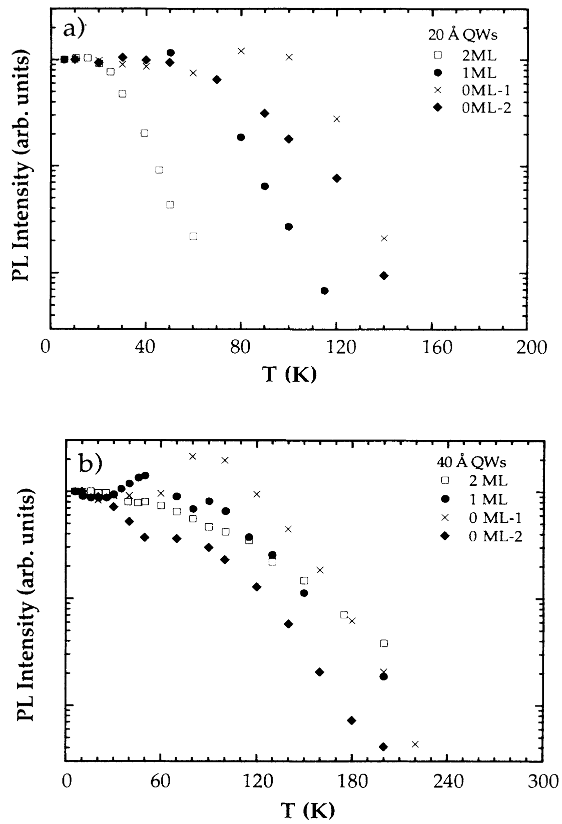

FIG. 3. Temperature dependence of the PL-integrated intensity I_L of the wells investigated: (a) 20-Å QW's; (b) 40-Å QW's.

pends on the well width and, in the case of the thinnest DBQW's, on the number of AlAs monolayers, decreases by several orders of magnitude, eventually reaching the time resolution of our experimental apparatus. It is well known that the low-temperature behavior of T_L mainly reflects the increase of the radiative recombination time;[12,13] on the contrary, it has been demonstrated that the high-temperature behavior of T_L, and therefore its decrease, is connected to the activation of some nonradiative channel.[13] We will show in the following that this process corresponds to the thermal escape of carriers out of the QW's.

In Fig. 3 we report the temperature dependence of the PL-integrated intensity relative to the recombination from the different QW's. In agreement with other reports,[8,9,13] we find a strong thermal quenching of the PL efficiency, again denoting the relevance of the nonradiative losses at high temperature. An anomalous behavior is observed in the PL intensity of the 40-Å QW's, where I_L tends to increase in correspondence of the decreasing of the PL intensity of the 20-Å wells. According to Ref. 9, we believe that this behavior can be explained in terms of a partial trapping, by the larger wells, of the carriers escaped out of the 20-Å QW's.

III. DISCUSSION

Let us first discuss the relationships between the measured quantities, namely the PL decay time T_L and the PL-integrated intensity I_L, and the radiative and nonradiative time constants, T_R and T_{NR}, respectively. In the case of time-resolved measurements, we obviously have

$$T_L = \frac{T_R T_{NR}}{T_R + T_{NR}} , \tag{1}$$

$$I_L = \eta_{exp} n N_0 \frac{T_{NR}}{T_R + T_{NR}} , \tag{2}$$

where η_{exp} stands for the experimental efficiency, N_0 is the number of carriers photogenerated by each laser pulse which are trapped by the QW, and n is the number of pulses inside the integration time. For continuous-wave (CW) excitation and steady-state condition, nN_0 obviously becomes the generation rate times the integration time. In principle, from Eqs. (1) and (2), one can directly extract both T_R and T_{NR}. The major difficulty in following this procedure comes from the evaluation of η_{exp} and N_0 with enough accuracy. In fact the temperature behavior of T_{NR} depends quite strongly on η_{exp} and N_0, while the effect on the radiative time T_R is only a scale factor.[13] Therefore we will concentrate our attention onto the limit of interest, namely very efficient nonradiative losses ($T_R \gg T_{NR}$). This assumption certainly holds, as it has been shown in Ref. 13, in the temperature region where the PL decay time of the QW's decreases with increasing the temperature. We get, in this region

$$T_l \cong T_{NR} , \tag{3}$$

$$I_L \cong \eta_{exp} n N_0 \frac{T_{NR}}{T_R} . \tag{4}$$

Hence the PL decay time coincides with the nonradiative time constant and the temperature dependence of T_{NR} is directly provided by the measurement of T_L. On the contrary, Eq. (4) shows that the temperature behavior of I_L is related to both T_R and T_{NR}. Only in the limit of negligible temperature dependence of T_R, the Arrhenius plot of the integrated PL intensity can allow to extract the activation energies of the nonradiative channels. This assumption is usually supposed to be valid for impurities[14] or color centers[15] but it certainly does not hold for quantum-well systems, due to the well-known temperature dependence of T_R.[12,13] In any case, it can be experimentally checked by the temperature dependence of the ratio T_L/I_L, which is proportional to T_R. In fact, as shown in Fig. 4, where the radiative time T_R is reported assuming negligible nonradiative losses at $T=4$ K [$T_R(4\text{ K})=T_L(4\text{ K})$], we find a strong temperature dependence of such ratio in agreement with Ref. 13.

It is also worth noting that, if T_R increases with increasing temperature, the measurement of the activation energy from the Arrhenius plot of the PL-integrated intensity leads to an overestimate of this quantity. Finally, even in the case of a constant radiative time T_R, Eqs. (3) and (4) show that the measurement of the PL intensity is an indirect method for obtaining information on T_{NR}. In particular, care has to be taken in order to keep η_{exp} and N_0 fixed when varying the temperature. Due to the temperature dependence of the band gap, both the PL recombination energy and the absorption coefficient will vary when heating the sample. Moreover, as it has been stressed very recently in Ref. 16, N_0 also depends on the trapping mechanism which can be temperature dependent.

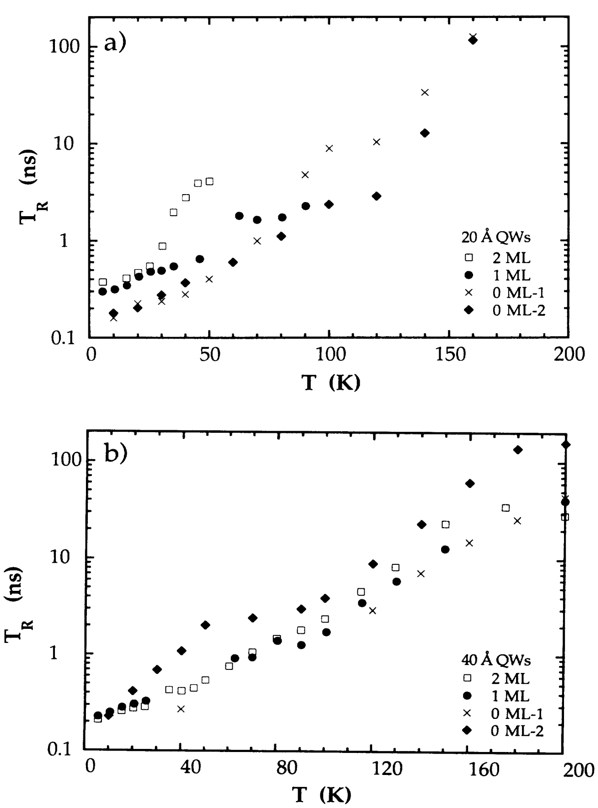

FIG. 4. Temperature dependence of the radiative time constant T_R of the wells investigated, chosing negligible nonradiative losses at $T=4$ K [T_R (4 K)$=T_L$ (4 K)]: (a) 20-Å QW's; (b) 40-Å QW's.

We now consider the effects on the recombination kinetics of the thermal escape of carriers out of the quantum well. Let us assume the carriers to be at thermal equilibrium at the lattice temperature. The whole PL band, which contains both exciton and free-carrier recombinations,[13,17] decays with a single time constant T_L given by Eq. (1), where T_R (T_{NR}) is the effective radiative (nonradiative) time coming from the average of the intrinsic recombination rates of each transition, weighted by the corresponding thermal populations.[18] We furthermore suppose that the main contribution to the nonradiative processes arises from thermal escape out of the well, under the assumption of a very efficient scattering rate Γ_0 from the confined states, having total energy larger than the barrier potential W_0, to the continuum. The three-dimensional continuum of states acts as a sink and we neglect the retrapping of carriers by the QW's. We thus have, in the nondegenerate case,

$$\frac{1}{T_{NR}} = \frac{\int_{W_0}^{\infty} dE\, \rho_{2D}(E)\Gamma_0(E)\exp\{-E/KT\}}{\int dE\, \rho_{2D}(E)\exp\{-E/KT\}} , \tag{5}$$

where $\rho_{2D}(E)$ is the two-dimensional (2D) density of states of the confined levels. Assuming a constant scattering rate Γ_0, isotropic and parabolic bands, and N_b bound states in the QW of energy E_n ($n=1,N_b$), we obtain

$$T_{NR} = \frac{\sum_{n=1}^{N_b} \exp\{-(En-E1)/KT\}}{N_b\Gamma_0\exp\{-\Delta/KT\}} , \tag{6}$$

where $\Delta = W_0 - E_1$ is the confinement energy of the ground state. For thin wells, having only one confined level, this result coincides with the time constant for the thermal ionization in the framework of a two-level model[15]

$$T_{NR} = \frac{1}{\Gamma_0\exp\{-\Delta/KT\}} . \tag{7}$$

It should be noted that the model is extremely simplified and several well-known features of the $GaAs/Al_xGa_{1-x}As$ system have been ignored, for instance the subband warping and nonparabolicity. The aim of this paper is indeed to stress the main features of thermal escape out of the QW's and we have chosen, therefore, to avoid unessential complications.

Combining Eqs. (3) and (6) we find that the slope of $\ln(T_L)$ versus $1/KT$, for high temperatures, leads to the characteristic confinement energy Δ of the kind of carriers escaping out of the well. Actually we expect the unipolar detrapping to be more efficient than the bipolar (or excitonic) one. Indeed, on one hand, it has been demonstrated that exciton ionization plays an important role in the kinetics of carrier recombination; in fact, excitons and free carriers are found to be in thermal equilibrium at the lattice temperature[13,17,19] with relative populations given by the 2D law of mass action. On the other hand, the carrier escape energy is less or equal to one half of the electron-hole-pair confinement energy. It should be also considered that the unipolar detrapping produces a spatial charge accumulation and therefore a band bending which, in principle, will oppose further carrier leakage into the barriers. However, this effect is negligible in our case, due to the small excess of photogenerated carriers ($\approx 10^{-10}$ cm^{-2}) at the excitation power used in our experiments.

In Fig. 5 we report the Arrhenius plot of $\ln(T_L)$ for the quantum wells investigated together with the fits of the high-temperature slope from which we directly obtain the scattering rate Γ_0 and the thermal activation energy E_A. As expected, the activation energies strongly depend on the well width and, in the case of the thinnest wells, on the number of AlAs monolayers. The scattering time $1/\Gamma_0$ comes out to be in the range 0.3–5 ps; we believe

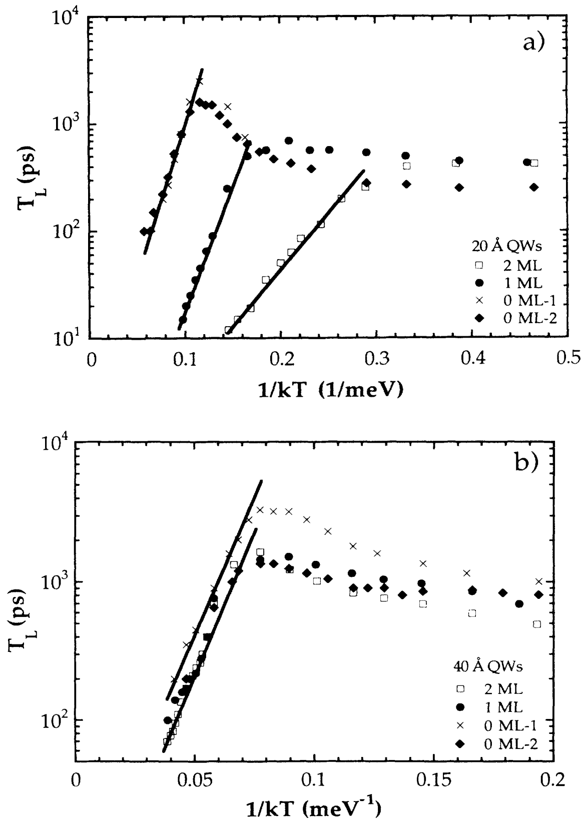

FIG. 5. Arrhenius plot of the PL decay time T_L versus $1/KT$ of the wells investigated: (a) 20-Å QW's; (b) 40-Å QW's. The straight lines are the best fits to the high-temperature slopes from which we have extracted the activation energies E_A reported in Table I. In (b) are reported only the fits to samples 0 ML-1 and 2 ML.

these values very reasonable, especially if one takes into account the simplicity of the model.

The activation energy E_A has to be compared with the carrier confinement energy Δ. From the experimental values of the band-gap discontinuity ΔE_g, fixing a given band offset ratio ΔE_c:ΔE_v, the heights of the potential barriers have been calculated for each sample. At the same time, the carrier confinement energies have been estimated by fitting the electron-hole-pair confinement energy $\Delta_{e\text{-}h}$ with a standard effective-mass model.[11] Unfortunately, even for a well-studied system like GaAs/Al_xGa_{1-x}As, several key parameters are not yet known with enough accuracy; in particular, we refer to the band offset ratio ΔE_c:ΔE_v which is usually assumed in the range between 60:40 and 70:30. This uncertainty is not relevant for analyzing the optical spectra which only measure the confinement energy of the electron-hole pairs, but becomes very important for determining the confinement energy of each carrier species.

In Table I we report the value of the activation energy E_A obtained from the fits to T_L and the confinement energies $\Delta_{e\text{-}h}, \Delta_e, \Delta_h$ of the electron-hole pairs, electrons, and holes, respectively. The last ones have been evaluated for two different band offset ratio ΔE_c:ΔE_v, i.e., 65:35 and 70:30, using an effective-mass calculation which takes explicitly into account the insertion of the AlAs thin cladding barriers, as reported in Refs. 10 and 11. We remark that in the calculations we have reduced, as usual, the nominal well widths by 5–10% in order to match the experimental energy positions of the PL peaks; obviously, different band offset ratios lead to slightly different well widths. Note also that in the case of the 40-Å QW's the activation energies come out from the values of T_L near room temperature where the PL intensity has dropped to very low levels, thus making the PL decay times affected by sizable errors. The reported values of E_A for the 40-Å wells have therefore to be considered an estimate, only, of the activation energies.

Referring to Table I, we see that in the case of the 2 ML DBQW of 20 Å the electrons have shallower subbands due to the peculiar characteristics of thin double-barrier quantum wells.[11] The electron and hole confinement energies become comparable in the case of the 1 ML DBQW of 20 Å and finally the electrons are more confined than the holes in all other wells investigated.

The activation energies found are much lower than the electron-hole confinement energies and in fair agreement with the escape out of the quantum wells of the less-confined carrier species; the agreement becomes better if a band offset 70:30 is chosen. We can therefore conclude that, at least in the samples investigated, i.e., thin GaAs/Al_xGa_{1-x}As QW's, the unipolar detrapping is the mechanism responsible for the thermal quenching of the QW photoluminescence. This is in disagreement with previous findings where the electron-hole escape has been claimed as the main loss mechanism on the basis of an analysis of the temperature behavior of the PL intensity.[8,9] We have demonstrated that this procedure is not correct if the radiative time constant T_R depends on the temperature, and that, for QW systems where T_R increases with increasing temperature, it leads to an overestimate of the activation energy.

IV. SUMMARY AND CONCLUSIONS

In summary, we have shown that the temperature dependence of the photoluminescence decay time T_L leads to the most direct measurement of the thermal activation of the nonradiative channels. The standard method based on the analysis of the integrated PL intensity can be used only if the temperature dependence of the radiative time constant T_R is negligible and therefore is not useful for the QW systems. From the slope of the Arrhenius plot of $\ln(T_L)$ versus $1/KT$ we have obtained activation energies in agreement with the unipolar escape of carriers out of the quantum wells instead of the bipolar one. We also find that a band offset ratio $\Delta E_c:\Delta E_v=70:30$ gives a better agreement with the experimental data than the commonly used value 65:35.

ACKNOWLEDGMENTS

One of us (J.M.P.) acknowledges financial supports from the Conselleria de Cultura, Educación i Ciencia de la Generalitat de València and from the P.F.P.I. del Ministerio de Educación y Ciencia. Work at CNRS-Sophia Antipolis is partially supported by the Commission of European Communities (Contract No. ST2J-254-3F).

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