Temperature dependence of the radiative and nonradiative recombination time in $GaAs/Al_xGa_{1-x}As$ quantum-well structures

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We report an experimental study of the photoluminescence (PL) properties, after both picosecond and continuous-wave excitation, of $GaAs/Al_xGa_{1-x}As$ quantum-well structures. The comparison between the PL decay time and the PL integrated intensity allows us to determine the temperature dependence of both the radiative and the nonradiative time constants. We find that the nonradiative processes play an important role and become dominant for $T \geq 100$ K. The radiative time constant turns out to be definitely higher than the measured PL decay time and increases by several orders of magnitude as the temperature is raised from $T=4$ K to room temperature. The experimental results are finally compared with the theories existing in the literature.

I. INTRODUCTION

The optical properties of quasi-two-dimensional electronic systems, such as the carriers photogenerated in quantum-well (QW) structures by optical absorption, have received increasing attention in recent years. In fact, the confinement of electrons and holes inside the QW's produces a strong enhancement of the excitonic effects[1] and gives rise to high optical nonlinearities[2] of crucial importance for device applications.

Despite the large number of studies dealing with carrier recombination in QW structures, no general agreement has yet been reached on basic questions such as the microscopic mechanisms involved in carrier recombination at a given temperature or the relevance of nonradiative-recombination channels.

On one hand, in fact, exciton recombination has been indicated as the main decay channel in QW's up to room temperature,[3] unlike in most bulk semiconductors. Furthermore, the increase of the photoluminescence (PL) decay time with temperature has been interpreted,[4,5] following Feldmann *et al.*,[4] as an evidence of radiative recombination of free excitons under the main assumption that the nonradiative processes play a negligible role.

On the other hand, it has been pointed out that the ionization of excitons is a very efficient process[6] which has to be taken into account even if the PL peak corresponds to exciton recombination.[7] Very recently, models have been proposed[8,9] for the radiative recombination which consider the thermal equilibrium between excitons and free carriers and predict both a nonlinear increase of the time constant with temperature and a nonexponential behavior of the radiative recombination. The conclusion has been drawn,[8] therefore, that the experimental results, which indeed show an exponential decay of the PL intensity even at room temperature, are controlled by nonradiative channels and that surface recombination gives, most likely, the major contribution.

In this paper, we present a detailed investigation of the temperature dependence of the PL decay time, after ps excitation, in a large set of $GaAs/Al_xGa_{1-x}As$ quantum wells grown by molecular beam epitaxy (MBE). We show that an increase, with increasing temperature, of the PL decay time is observed in all samples, followed by a rapid decrease starting from a value of the temperature depending on the well width: the narrower the well is, the lower is the temperature at which the PL decay time reaches the maximum value.

A combined analysis of the temperature dependence of the PL decay time and the integrated PL intensity allows us to separate the radiative and nonradiative recombination times. In particular, it is found that the radiative recombination time increases with temperature much faster than the experimental values of PL decay time and that the nonradiative processes indeed play an important role up to dominate the recombination at high temperature.

Furthermore, the analysis of the spectral features of the PL, carried out by means of continuous-wave (CW) photoluminescence and photoluminescence excitation (PLE) spectra, makes it possible to identify the different contributions to the radiative recombination such as bound excitons, heavy and light free excitons and free carriers with a relative weight depending on the value of the temperature.

Finally, the radiative recombination time, extracted

from the experimental data, is compared with the predictions of the models proposed in the literature for the radiative recombination in quantum wells. We find that our results are outside any models dealing with exciton recombination alone, and only in a poor qualitative agreement with a model[9] based on the thermal equilibrium between excitons and free carriers. A discussion of the possible open problems in both the experimental and theoretical analyses is also given.

The paper is organized as follows. Details on the structure and growth characteristics of the samples investigated, together with a brief description of the experimental setup, is given in Sec. II. In Sec. III we report the experimental data of both CW and time-resolved measurements. A discussion of the results and the models proposed in the literature is given in Sec. IV. A summary of the main results and the conclusions are reported in Sec. V.

II. SAMPLES AND EXPERIMENT

All the samples investigated were nominally undoped $GaAs/Al_xGa_{1-x}As$ QW's grown by MBE on a (001) oriented semi-insulating GaAs substrate kept at a temperature of about 600 °C, with growth interruption (GI) at both interfaces in most cases. Most of the samples consisted of three QW's of different widths, ranging from 20 to 150 Å, in between barriers thick enough (200–300 Å) to completely separate the confined levels inside each well. This kind of structure was chosen in order to investigate the optical properties of QW's with different thicknesses grown under identical conditions so as to have similar extrinsic characteristics. Two samples had three identical QW's of 70 Å width and a last sample was a single quantum well 40 Å thick. A 0.5-μm GaAs buffer layer was always grown before the active structure. We report in Table I the list of the samples investigated together with the nominal well and barrier thicknesses, aluminium content, and whether or not interruption has been used during growth.

PL spectra under CW excitation have been taken using a He-Ne or an Ar^+ laser, while an argon-pumped Ti:sapphire laser has been used as excitation source in PLE measurements. The PL signal was dispersed through a 0.6-m double monochromator and detected, by a standard photon counting technique, using a cooled GaAs photomultiplier.

Time-resolved PL measurements have been performed using a synchronously pumped mode-locked dye laser; the time duration of the pulses was 3 ps, with a repetition rate of 76 MHz and the wavelength tuned to $\lambda_{exc}=6100$ Å. The luminescence was analyzed by a time-correlated single photon counting apparatus providing a time resolution of the order of 70 ps, by using standard numerical deconvolution. 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.

III. EXPERIMENTAL RESULTS

A. Continuous-wave measurements

A comparison between typical PLE and PL spectra at different temperatures for a 70-Å well is shown in Fig. 1. At low temperature, $T=4$ K [Fig. 1(a)], we find a Stokes shift of about 3 meV between the PL and PLE peaks, indicating exciton localization at crystal defects. Increasing the temperature, the resonances in the PL and PLE spectra shift, as expected, towards lower energies, while, at the same time, the energy difference between the PL and PLE peaks decreases; finally, for $T=50-70$ K, depending on the sample, the Stokes shift vanishes [Fig. 1(b)], suggesting[10] that free-exciton recombination becomes dominant. For higher values of the temperature, the PL intensity decreases by orders of magnitude thus making very difficult to perform PLE spectra for $T\geq 200$ K; we still find, at this temperature, as reported in Fig. 1(c), the PL and PLE peaks coincident at the same energy position.

On the other hand, sharp resonances are present in the PL spectra up to room temperature; reporting in a graph the energy position as a function of the temperature, we find a good agreement with the T dependence of the GaAs band gap[11] in the range 60–300 K. This allows us to assign the peaks in the PL spectra to free-exciton recombination up to room temperature.

Similar features have been observed in all the samples investigated; the Stokes shift is in the range 1–8 meV and the full widths at half maximum of the PL lines at low temperature are between 3 and 9 meV, indicating a good

TABLE I. Table of the samples investigated. We report the well width (L_{WJ}; $J=1,2,3$), the barrier width (L_B), the aluminium content (X), and the growth interruption (GI), if applied.

Sample No.	L_{W1} (Å)	L_{W2} (Å)	L_{W3} (Å)	L_B (Å)	X	GI
1	70	70	70	200	0.30	no
2	70	70	70	200	0.30	yes
3	50	70	150	300	0.30	no
4	50	70	150	300	0.30	yes
5	20	40	80	300	0.32	yes
6	20	40	80	300	0.32	yes
7	40			420	0.28	yes

overall quality of all samples.

The analysis of PL and PLE spectrum line shapes provides us with additional useful information on the radiative recombination mechanisms in these structures. The shoulder at higher energies with respect to the first heavy exciton state E_{1H}, which becomes the more evident the higher the temperature is, can be unambiguously assigned to the light exciton recombination, as it is clearly evident from a comparison with the PLE spectra (see, for instance, Fig. 1) and in agreement with theoretical predictions based on standard effective mass models.

Furthermore, the presence of a thermal tail extending for several tens of meV [Fig.1(c)] at the high-energy side of the emission peaks and having a slope in agreement with the nominal temperature of the sample is evident for $T \geq 70-100$ K. In a recent paper[7] it has been demonstrated, by means of a detailed fitting of the PL line shape, that this tail is originated by recombination of free carriers generated from exciton ionization. Moreover, the fits allowed us to determine the relative populations of excitons and free carriers as a function of temperature

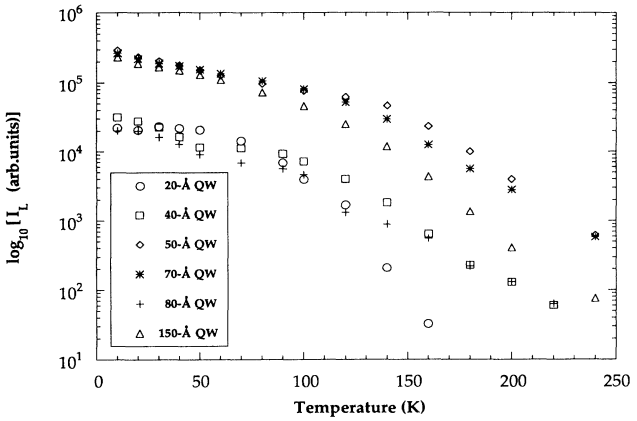

FIG. 2. Temperature dependence of the PL intensity, integrated over the whole PL band, from QW's of thickness varying from 20 to 150 Å (sample nos. 6 and 3).

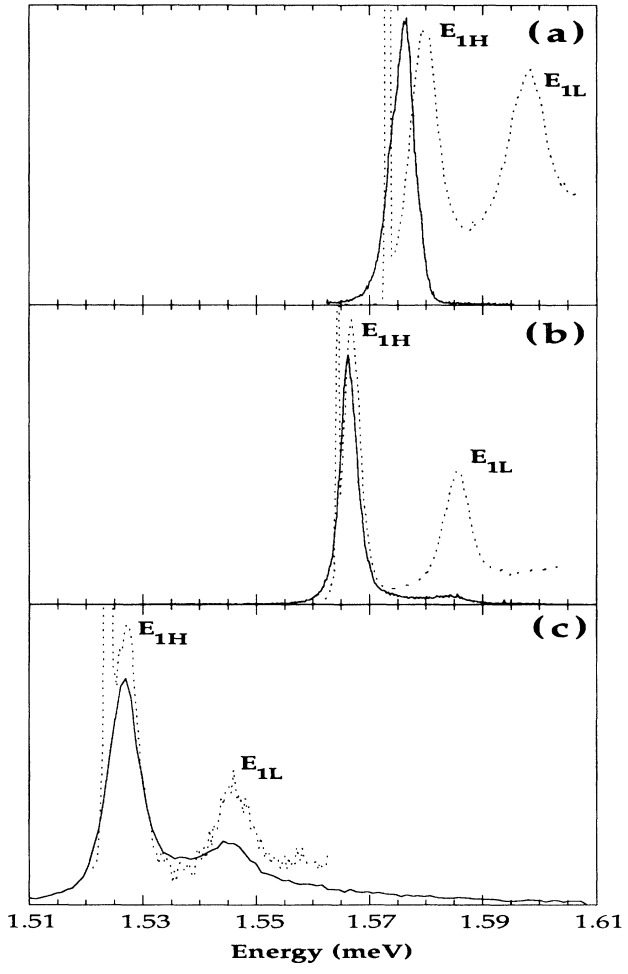

FIG. 1. Comparison between the PL (solid line) and PLE (dotted line) spectra at (a) $T=4$ K, (b) $T=80$ K, (c) $T=200$ K in the case of a 70-Å well (sample no. 1). The heavy and light exciton lines have been labeled by E_{1H} and E_{1L}, respectively. Note the absence of Stokes shift at high temperatures.

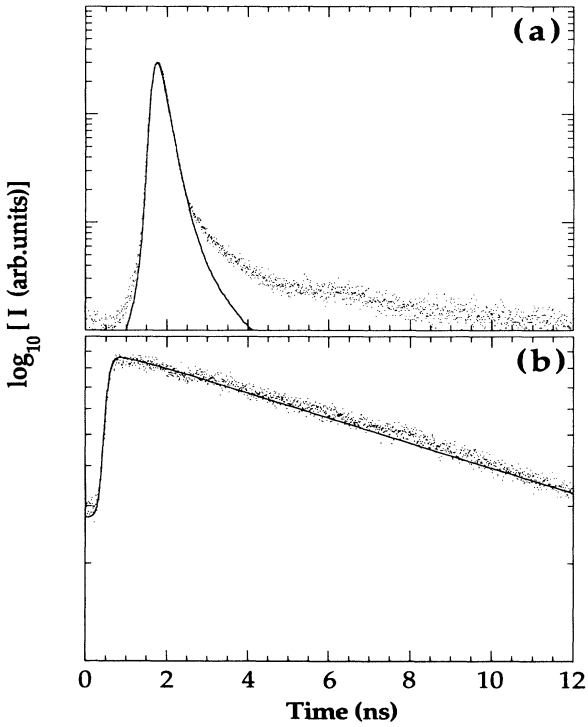

FIG. 3. Typical PL decay curves (dots) at two different temperatures, (a) $T=4$ K and (b) $T=150$ K, for a 70-Å well (sample no. 3). The solid curves give the fit to the experimental points assuming a monoexponential decay, after convolution with the instrumental response function.

and to show the validity of the two-dimensional (2D) law of mass action in this temperature range.

The temperature dependence of the PL-integrated intensity relative to the emission from the different QW's, from liquid helium up to room temperature, is reported in Fig. 2. We find, in all the samples investigated, a decrease of the PL radiative efficiency by several orders of magnitude, in agreement with the finding of Chen *et al.*[12] This general feature does not depend, apart from minor differences, on the excitation wavelength used, even when it is changed from very high in the continuum ($\lambda_{exc}=5145$ Å) to below the band gap of the $Al_xGa_{1-x}As$ barrier ($\lambda_{exc}=7300$ Å); this fact clearly suggests that the strong decrease of PL intensity with temperature is mainly connected with the dynamics of the states involved in the radiative recombination.

B. Time-resolved measurements

The dependence of the photoluminescence emission intensity at the heavy exciton energy position E_{1H} as a

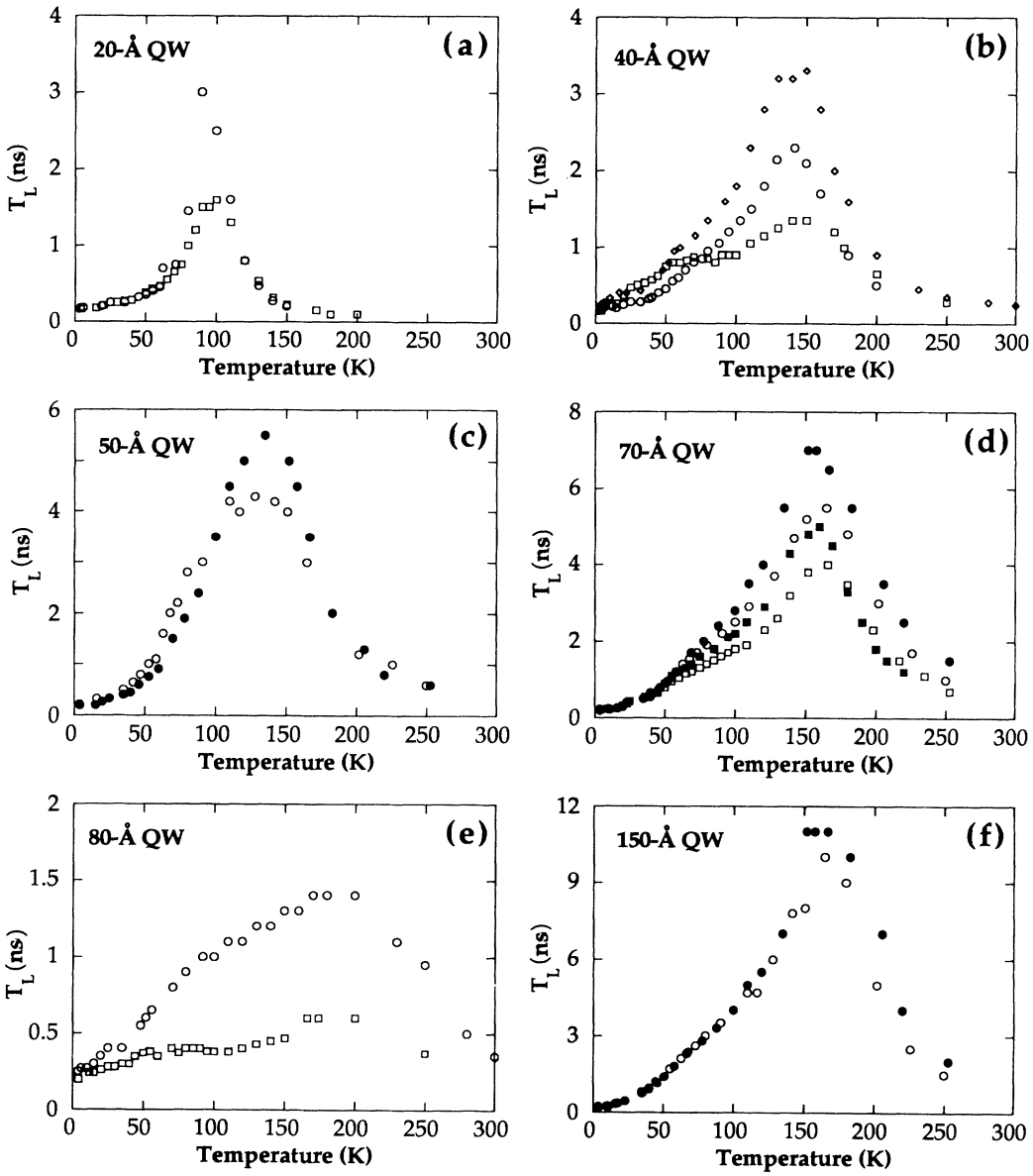

FIG. 4. Temperature dependence of the PL decay time T_L for different well thicknesses. Different symbols in the same figure refer to different samples having the same well width: (a), (b), (e) circles: sample no. 5; squares: sample no. 6; (b) rhombi: sample no. 7; (c), (d), (f) solid (open) circles: sample no. 3 (4); (d) solid (open) squares: sample no. 1 (2). Note in (c), (d), and (f) the comparison between identical samples with (open symbols) and without (solid symbols) growth interruption at the interfaces.

function of time after the excitation pulse is shown in Fig. 3 for a 70-Å well, at two temperatures, together with the fits obtained by assuming a monoexponential decay and convolving for the instrumental response function.

We find that the experimental data are consistent with a pure exponential decay with time constant T_L over, at least, 1 order of magnitude even if, at low temperature, $T \leq 50$ K, monoexponential fits are of a poorer overall quality. This is connected with the fact that, as discussed below when reporting the time-resolved PL spectra, a temporal dynamics is observed, at low temperatures, inside the exciton band: different spectral components decay with different lifetimes and therefore a multiexponential fit[13] would be more adequate for describing the PL decay. At higher temperatures, instead, a single time constant is observed in the whole band.

The temperature dependence of the PL decay times T_L, reported in Fig. 4, is similar for all the QW's investigated. At liquid-helium temperature ($T=4$ K) the value of T_L is, within the experimental accuracy (~ 50 ps), almost independent on the QW thickness and results in the order of 250 ps. Raising the temperature T_L increases in a nonlinear way with a change of slope around 50–70 K; then, after reaching a maximum value of several ns, decreases rapidly to hundreds of ps when approaching room temperature.

Several features have to be stressed. First of all, the maximum value of T_L is strongly sample dependent; in fact, we find up to a factor of 3 between QW's having identical design parameters. Secondly, apart from this inhomogeneity, the value of the temperature at which such maximum is reached seems to be an intrinsic property of the QW thickness; in fact, it turns out to be the same in different samples having the same nominal structure. It is also worth noting that, for narrow wells, the maximum value of T_L is reached at lower temperature than in wider wells. Finally, as far as the effect of growth interruption at the interfaces is concerned [Figs. 4(c),

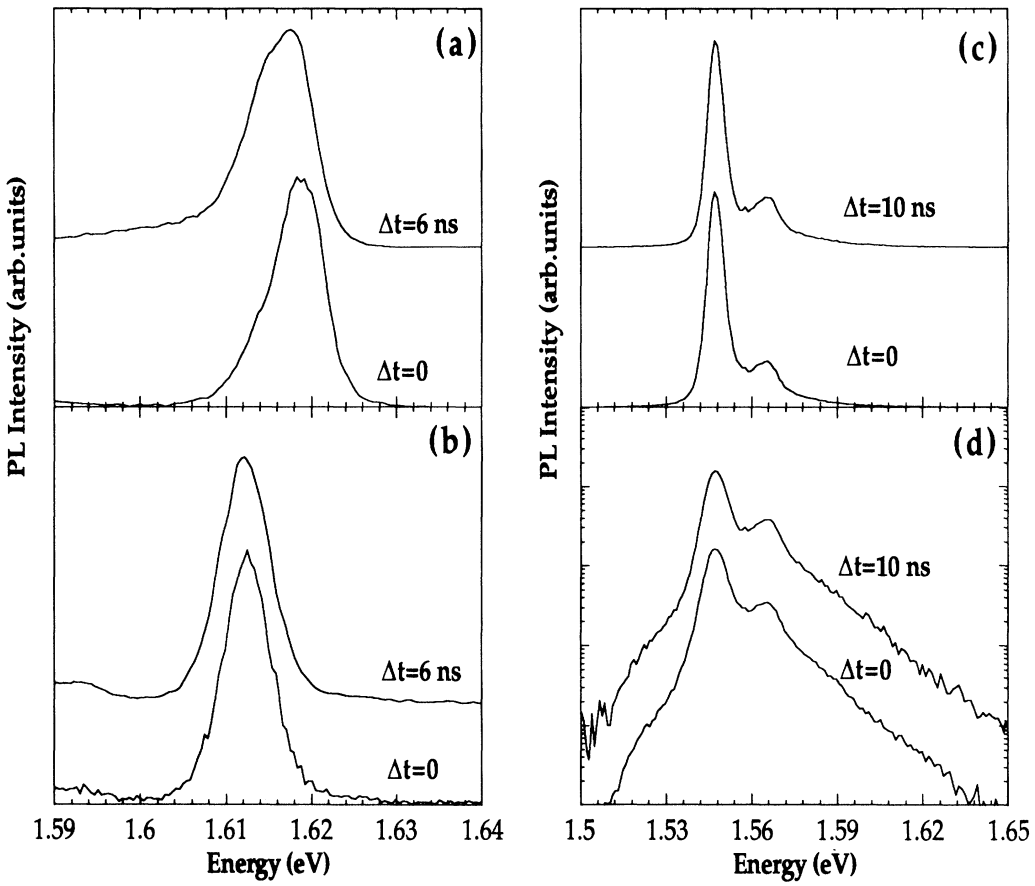

FIG. 5. Typical time-resolved spectra for different delays Δt after ps excitation. (a) 50-Å well (sample no. 3), $T=4$ K; (b) 50-Å well (sample no. 3), $T=70$ K; (c) 70-Å well (sample no. 1), $T=150$ K; (d) semilogarithmic plot of the same spectra as in (c). Note the temporal dynamics in the time-resolved spectra at $T=4$ K: the peaks shift towards lower energies and the FWHM increases with Δt. On the contrary, the whole PL band decays with only one time constant at high temperatures, thus indicating the thermal equilibrium between different exciton states and free carriers.

4(d), and 4(f) for the 50-, 70-, and 150-Å QW's], we find a decrease of the PL decay time associated with the interruption during the growth, in qualitative agreement with Ref. 14, where, however, the effect of GI is more pronounced.

Typical time-resolved spectra are shown in Fig. 5 for different temperatures and different delay times Δt after excitation. As already noted, we find that the spectra at low temperatures [$T \leq 50$ K; see, for instance, Fig. 5(a)] show a temporal dynamics inside the emission band; the PL peak shifts, with increasing time delay after the excitation pulse, towards lower energies while the FWHM increases. On the contrary, no temporal dynamics is observed at temperatures higher than 60–70 K despite the fact that several different contributions, namely, recombination of heavy/light excitons and free carrier recombination, can be resolved in the PL spectra. It is worth noting that we measure the same lifetime over a PL band spanning over more than 100 meV [Figs. 5(c) and 5(d)]; as we will discuss later, this is a direct indication that the carriers are at thermal equilibrium.

IV. DISCUSSION

In this section we discuss the implications of the whole set of measurements presented in Sec. III on the mechanisms involved in carrier recombination, at a given temperature, in $GaAs/Al_xGa_{1-x}As$ quantum wells. We recall, first of all, that the measured time decay T_L at temperature T is related to the radiative and nonradiative recombination times T_R and T_{NR}, respectively, by

$$\frac{1}{T_L(T)}=\frac{1}{T_R(T)}+\frac{1}{T_{NR}(T)} . \tag{1}$$

We will suppose, in the following, that the carriers in the levels which undergo recombination are at thermal equilibrium; in other words, the thermalization processes are much faster than the recombination processes. This assumption implies a strong coupling between these states so that a single time constant will describe the radiative processes inside the whole PL band; the same holds for the nonradiative processes.

At the same time, the temperature dependence of the luminescence intensity $I_L(T)$ integrated over the whole PL band can be expressed as

$$I_L(T)=I_0\frac{T_L(T)}{T_R(T)}=I_0\eta(T) , \tag{2}$$

where $\eta(T)$ is the radiative efficiency at temperature T and I_0 is a normalization factor which depends on the number of photoexcited carriers. Even if the combined measurement of PL decay time and intensity is a standard method for determining the radiative and nonradiative recombination times,[15,16] we want to remark that relation (2) is correct under the main assumption that the efficiency of the relaxation processes, from the level of excitation to the states that undergo radiative recombination, does not depend on temperature. As stressed before, this assumption is supported by the observation that the PL intensity $I_L(T)$ shows a similar dependence on T irrespective of the different relaxation paths followed by the photogenerated carriers when different excitation energies are used.

It follows from Eqs. (1) and (2) that

$$T_R(T)=I_0\frac{T_L(T)}{I_L(T)}=\frac{T_L(T)}{\eta(T)} , \tag{3}$$

$$T_{NR}(T)=T_L(T)\frac{I_0}{I_0-I_L(T)}=T_L(T)\frac{1}{1-\eta(T)} . \tag{4}$$

We can therefore extract both the radiative and nonradiative recombination times, as a function of T, from the combined measurements of $T_L(T)$ and $I_L(T)$.

Typical results for the temperature dependence of $T_R(T)$ and $T_{NR}(T)$ are reported in Figs. 6 and 7 under two different assumptions for the radiative efficiency η at liquid-helium temperature, namely, $\eta(T=4\text{ K})=0.9$ and $\eta(T=4\text{ K})=0.5$, respectively. Note that, while the temperature dependence of $T_R(T)$ does not change, apart from a scale factor, when assuming different values for the normalization constant I_0, the behavior of $T_{NR}(T)$ as a function of T is rather sensitive, particularly at low temperatures, to the chosen values of I_0 or, equivalently, to the absolute radiative efficiency $\eta(T)$ at the given temperature.

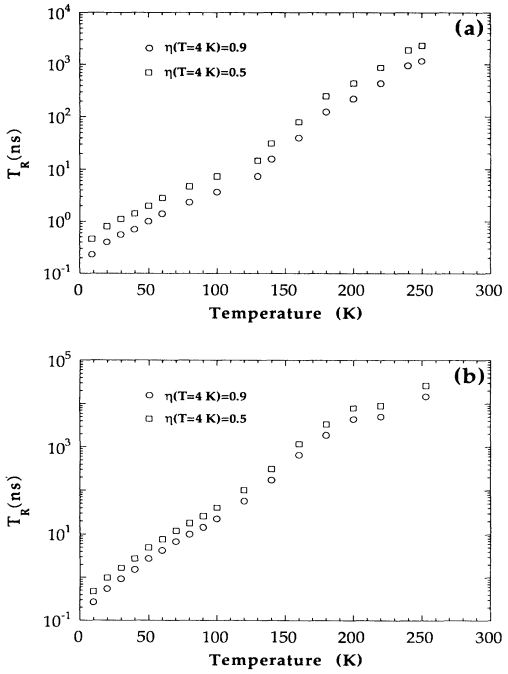

FIG. 6. Temperature dependence of the radiative recombination time extracted from the combined measurements of $T_L(T)$ and $I_L(T)$. (a) 70-Å well (sample no. 1), (b) 150-Å well (sample no. 4). The two sets of points in each frame correspond to different values for the radiative efficiency at 4 K, namely, $\eta(T=4\text{ K})=0.9$ (circles) and $\eta(T=4\text{ K})=0.5$ (squares).

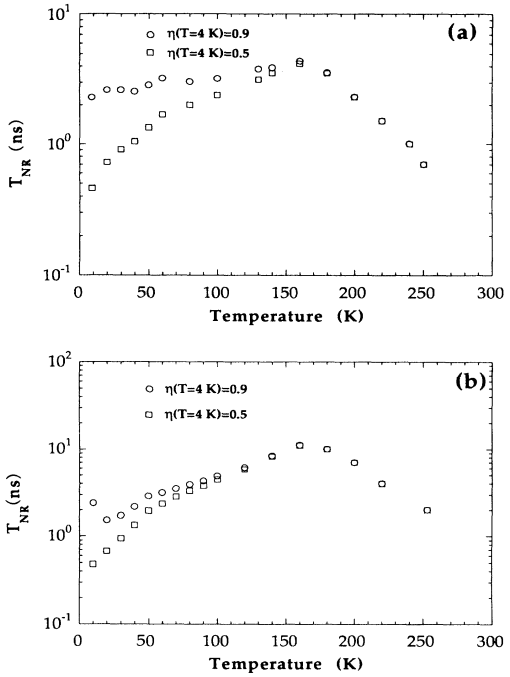

FIG. 7. Temperature dependence of the nonradiative recombination time extracted from the combined measurements of $T_L(T)$ and $I_L(T)$. (a) 70-Å well (sample no. 1), (b) 150-Å well (sample no. 4). The two sets of points in each frame correspond to $\eta(T=4\text{ K})=0.9$ (circles) and $\eta(T=4\text{ K})=0.5$ (squares).

We see that the decrease of the PL-integrated intensity is associated with a strong increase of the radiative recombination time $T_R(T)$; at the same time the behavior of the PL decay time $T_L(T)$ turns out to be closely connected with the main features of the nonradiative recombination time $T_{NR}(T)$. We find that $T_R(T)$ increases with T much faster than linearly in all samples and that $T_{NR}(T)$ gives a non-negligible contribution to the observed PL decay time $T_L(T)$ at all temperatures; the nonradiative decay channels dominate the recombination processes in our samples as soon as the temperature is higher than 100 K.

We would like to stress that the usual identification[4,5] of $T_L(T)$ with $T_R(T)$, in the absence of any comparison with the radiative efficiency, may therefore turn out incorrect. Let us now separately discuss the results obtained for $T_R(T)$ and $T_{NR}(T)$.

A. Radiative recombination time

A phenomenological model for the temperature dependence of the radiative recombination time of excitons in QW's has been proposed by Miller *et al.*[16] several years ago; a similar approach was then independently assumed by Feldmann *et al.* in a recent well-known paper.[4] In this picture, excitons can only recombine radiatively if their kinetic energy is less than a characteristic value Δ determined by the interaction with acoustic phonons. Therefore, the exciton states can be divided into two classes: radiative states, for $E<\Delta$, with a finite value of the lifetime $T_R(E)$ and nonradiative (surface polariton-like states), for $E>\Delta$, which cannot recombine into a photon [$T_R(E)=\infty$]. The thermal equilibrium inside the exciton band then determines the existence of only one radiative time constant for the whole band which comes out from a thermal average over the radiative and surface states. The result is a linear increase of T_R with temperature for T higher than Δ/K_B, where K_B is the Boltzmann constant. On the other hand, the microscopic mechanism for the decay is not specified by the authors and the slope of the linear increase of T_R depends on phenomenological parameters.

A more refined theory for the radiative lifetime of free excitons has been recently presented by Andreani *et al.*[17] The theory relies on a microscopic polariton picture for the recombination of free excitons in QW's based on the conservation of the in-plane wave vector $K_{\parallel}$ and the instantaneous thermalization over the whole band. A detailed model, including the valence-band mixing and the coupling with the continuum levels, is used for calculating the exciton states and a radiative lifetime, as short as 25 ps, is predicted at $K_{\parallel}=0$ for the heavy exciton in $GaAs/Al_xGa_{1-x}As$ QW's of about 100 Å. The thermal equilibrium with surface states strongly increases the effective radiative time of the exciton band as soon as the temperature overcomes 1 K, resulting in a linear increase of $T_R(T)$ with a slope of 34 ps/K for a 100-Å wide $GaAs/Al_{0.3}Ga_{0.7}As$ QW.[17]

We would like to stress once more that the observed increase of the PL decay time with T has been interpreted, in many papers,[4,5] in the framework of the exciton model for T_R proposed by Feldmann *et al.*[4] We have already discussed the fundamental distinction between T_L and T_R and now we emphasize, again, that a large part of the experimental data presented in Sec. III are outside any model considering free-exciton recombination alone.

In fact, the Stokes shift observed between the PL and PLE peaks up to $T\approx 50$ K (see Fig. 1) is clear evidence that the recombination at low temperature occurs from localized excitons rather than free excitons. This can also explain the fact that, in our samples, the low-temperature value of T_L results to be almost independent on the well thickness, contrary to what is found in the case of recombination arising from free excitons, as claimed in Ref. 4. The peaks in the PL spectra can be ascribed to free-exciton recombination at the heavy valence subband only for $T\geq 50-70$ K in our samples; at the same time, clear evidence of exciton recombination at the light hole subband and recombination of free carriers is provided by our data.

Furthermore, the exponential tail at the high-energy side of the PL spectra, together with the time-resolved spectra of Fig. 5, support the effectiveness of exciton ionization.[7] A thermal equilibrium between excitons and free carriers is demonstrated, at least for $T\geq 70-100$ K, by the presence of only one decay constant inside the whole PL band.

Therefore, other mechanisms, besides heavy exciton recombination, are effective in the radiative process and the measured radiative time constant has to come out from a thermal average over the characteristic recombination constants of each component.

We conclude that the different theories on the temperature dependence of the radiative recombination time of excitons in QW's can only be tested in samples which do not show any Stokes shift at low temperature, and, in a small range of temperatures ($T \leq 70$ K), in order to avoid both the thermal population of higher subbands and the effects of exciton ionization.

Only very recently, models[8,9] have been proposed for the kinetics of the radiative recombination in QW structures which take into account the effects due to exciton ionization. The main assumptions of these theories are that exciton ionization is much faster than the radiative recombination time and that the thermal equilibrium between excitons and free carriers follows the law of mass action. It is also stressed in these papers that, given the fact that the radiative recombination of free carriers is a bimolecular process, the radiative time decay would turn out to be nonexponential[8,9] and dependent on the carrier density.[9]

We report in Fig. 8 a comparison of our data for $T_R(T)$ and the prediction of the model by Ridley[9] in the limit of low excitation density such as in the case of our experiment. We see that the radiative constant predicted by the model increases with temperature by approximately the correct orders of magnitude, even if the temperature dependence provided by the model does not follow the experimental behavior.

Let us discuss some possible reasons for the poor agreement between experimental data and theoretical predictions. First of all, Ridley's model[9] does not include experimental evidences such as the exciton localization at low temperature and the thermal population of the light exciton states; it also neglects the temperature dependence of the free carrier's recombination constant.[18] On the other hand, we find that the inclusion of the last two points does not significantly change the overall predictions, given the fact that the temperature dependence of $T_R(T)$ is dominated by the effects of exciton ionization. At the same time, the effects of the thermal transition from bound to free excitons has been studied in a recent paper[19] where it is shown that this process does not drastically modify the recombination time.

Secondly, the model of Ridley[9] assumes the 2D law of mass action to also be valid at very low temperature where we do not know of any experimental confirmation and where, moreover, our data show the existence of exciton localization.

As far as the experimental data are concerned, the main assumption underlying the determination of T_R is the commonly adopted hypothesis[15,16] that the PL-integrated intensity $I_L(T)$ coincides, apart from the constant I_0, with the inverse of the radiative efficiency $\eta(T)$; in other words, we suppose that the efficiency of the relaxation processes does not change with the temperature. This is confirmed, but not definitely proved, by the fact that only minor changes are observed, for the temperature dependence of $I_L(T)$, when varying the excitation energy.

A more refined measurement of $\eta(T)$ could be *a priori* obtained by performing a resonant excitation inside the PL band, so as to eliminate the relaxation processes. Unfortunately, due to the reduction of the absorbing volume, a strong decrease of the intensity is observed under resonant excitation, making it difficult to perform PL experiments at the higher temperatures. Furthermore, significant modification of the PL line shape,[20] together with a reduction in the PL efficiency,[20,21] can be observed in GaAs/$As_xGa_{1-x}As$ QW structures, after resonant excitation, possibly connected with the presence of deep levels in the barriers;[21] we refer to another paper[20] for the discussion of these points.

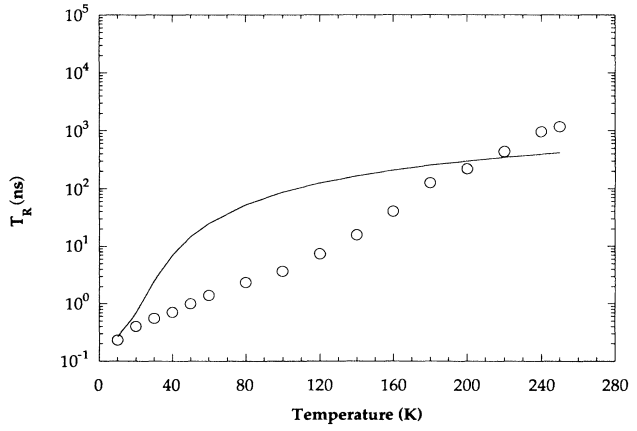

FIG. 8. Comparison of the radiative recombination time as obtained from a 70-Å well (sample no. 1) for $\eta(T=4\text{ K})=0.5$ with the prediction of the Ridley's model (Ref. 9) (solid line), adjusting for a scale factor in order to match the low-temperature values of T_R.

B. Nonradiative recombination time

As already discussed and reported in Fig. 7, while the temperature dependence of the radiative recombination time T_R does not depend on the normalization value chosen for the absolute radiative efficiency η, the temperature dependence of the nonradiative recombination time T_{NR} turns out to be rather sensitive to the value of η at least for $T \leq 100$ K. At higher temperatures, $T_R \gg T_{NR}$ and the temperature dependence of the PL decay time reflects, to a large extent, the nonradiative processes ruling the recombination in our samples. This is consistent with the observation in Ref. 8 that, if the radiative processes were to dominate the recombination, one would find a nonexponential behavior for the PL time decay, contrary to the experimental observation.

Very little is known,[15,22] to our knowledge, about the main nonradiative decay processes occurring in

$GaAs/Al_xGa_{1-x}As$ QW's and from our set of data we can only extract some crude indications.

The peaking of T_L at a value of the temperature dependence on the well width might suggest, in our opinion, the importance of surface recombination and/or Shockley-Read recombination via states at the $GaAs/Al_xGa_{1-x}As$ interfaces, as, indeed, indicated by Pickin and David[8] to give the main nonradiative decay channel at room temperature in these structures. Unfortunately, we do not know of any result concerning the temperature dependence of the surface recombination velocity in QW's that might help in settling the question.

At the same time, the spread in the experimental values of T_L, at a given temperature, in QW's having the same nominal thickness seems to suggest a dependence of the nonradiative processes on extrinsic properties connected with the sample growth. We have found, for instance, that the growth interruption at the interfaces gives rise to slightly smaller values of the PL decay time if compared with similar samples grown without GI. This is in qualitative agreement with the results of Ref. 14, where, however, the effect is much more pronounced and can be interpreted as an increase of trap and impurity incorporation associated with the growth interruptions.[14] A detailed study of the effects of the interruption during the growth on T_{NR} is in progress.

In a recent paper,[23] a comparison has been reported between the effects of interface defects and impurity incorporation on the PL properties of QW's showing the major role played by impurity contamination in reducing the PL efficiency and changing the characteristic recombination time. Carrier trapping at deep levels is therefore suggested as an efficient nonradiative process in these samples. Apart from these qualitative indications, further experimental work is clearly needed in order to reach a better understanding of the nonradiative processes ruling carrier recombination in QW's, particularly at high temperature.

V. SUMMARY AND CONCLUSIONS

We have presented a detailed investigation of the PL spectral features and time decay in $GaAs/Al_xGa_{x-1}As$ quantum-well structures. The combined measurements of the PL decay time and PL-integrated intensity as a function of the temperature have allowed us to determine both the radiative and nonradiative time constants, clearly showing the difference between the PL decay time T_L and the radiative recombination time T_R. We have found that T_R increases with temperature much faster than linearly in contrast with the well-known and accepted exciton model of Feldmann *et al.*;[4] in fact, our experimental data are outside any simple model involving only the first exciton band.

We have shown that a comparison between the PL and PLE spectra suggests that recombination at low temperatures ($T \leq 50$ K) occurs from an exciton localized at crystal defects while the peaks in the PL spectra correspond to free-exciton recombination only at higher temperatures. At the same time, as soon as $T \geq 70$ K, clear evidence of recombination of the light hole exciton and exciton ionization into free carriers has been given. We have also found that the 2D carrier system is in thermal equilibrium when the temperature is greater than 70 K so that the effective radiative recombination time has to come out from a statistical average of the intrinsic recombination times of each of the states involved in the recombination.

The model of Ridley,[9] based on the thermal equilibrium between excitons and free carriers, seems to provide a more reasonable framework for the discussion of the experimental data, even if the recombination from localized excitons observed at low temperature is still outside Ridley's model and the extrapolation down to very low temperatures of the 2D law of mass action for the exciton ionization is not confirmed by the analysis of the PL spectra. The comparison with the experimental data shows an unsatisfactory agreement; nevertheless, the model predicts an increase of $T_R(T)$, from 10 K up to room temperature, by the correct orders of magnitude.

The importance of the nonradiative decay channels has been stressed together with some crude indications of possible processes that might play a major role, namely, interface recombination and carrier trapping at deep levels. The lack of a satisfactory agreement between theory and experimental data suggests that further work, both theoretical and experimental, is needed in order to fully elucidate the main mechanisms underlying carrier recombination in $GaAs/Al_xGa_{1-x}As$ quantum-well structures.

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