Temperature dependence of exciton lifetimes in GaAs/$Al_xGa_{1-x}As$ single quantum wells

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We report on continuous-wave and time-resolved measurements of the photoluminescence of good-quality single quantum wells at low temperatures (4–30 K). Luminescence arising from both free and localized excitons is observed and the influence of excitation localization on the photoluminescence decay time is investigated. Resonant pumping at the light-hole exciton transition is found to greatly increase the generation of free heavy-hole excitons. In this case, the free-exciton lifetime increases linearly with temperature (10–30 K) and the dependence of the slope on the well thickness is found to be in good quantitative agreement with the theoretical model of Andreani *et al.* [Solid State Commun. 77, 641 (1991)].

I. INTRODUCTION

The linear and nonlinear properties of quantum wells (QW's) are dominated, to a large extent, by excitonic effects.[1] In fact, the increase of exciton binding energy[1–4] and oscillator strength[4,5] due to size quantization is responsible for the strong enhancement of the absorption and emission at the exciton transition energies, even at room temperature.

Unfortunately, intrinsic free-exciton properties are difficult to observe in photoluminescence (PL) experiments at low temperatures, because of the strong contribution arising from bound excitons whenever a sizable interface roughness and/or impurity concentration is present in the QW structure. As a consequence, exciton lifetimes measured by time-resolved photoluminescence (TRPL) turn out to be sample dependent, and a wide range of decay times (0.1–1 ns) can be found in the literature.[6–15] At the same time, no conclusive experimental evidence has been reported so far on the validity range of recent theoretical models for the radiative lifetime of free excitons in quantum wells and, in particular, its dependence on temperature.[4,7,16]

In this work, we report experimental results on the luminescence and lifetime of excitons in a set of high-quality GaAs/$Al_{0.3}Ga_{0.7}As$ single quantum wells, characterized by narrow PL lines and zero (or near-zero) Stokes shifts.

Continuous-wave (cw) and time-resolved experiments have been carried out in order to investigate the temperature dependence of the exciton lifetimes in these structures. The influence of the excitation energy and the effect of exciton localization at interface fluctuations on the measured lifetimes have been elucidated.

Finally, in the case of free-exciton recombination, the well-width dependence of the linear increase with temperature of the excitonic lifetimes has been found to be in agreement with the predictions of the model by Andreani and co-workers.[4]

II. SAMPLE AND EXPERIMENTAL SETUP

The sample investigated consists of five GaAs single quantum wells of different thickness (40, 60, 80, 120, and 180 Å) grown on a semi-insulating GaAs substrate with a (001) orientation. The QW's are separated from each other by 500-Å-thick $Al_{0.3}Ga_{0.7}As$ barriers.

Continuous-wave photoluminescence and PL excitation (PLE) measurements have been carried out using, for excitation, an Ar^+-pumped Ti:sapphire laser. The PL signal was dispersed by a double-grating monochromator (spectral resolution: 0.2 meV) and analyzed by standard photon-counting techniques.

The excitation source for TRPL measurements was a mode-locked Nd:YAG (yttrium aluminum garnet) synchronously pumped dye laser, providing 5-ps pulses in the range 700–820 nm at a repetition rate of 76 MHz. The PL signal was dispersed through a 0.22-m double monochromator (spectral resolution: 1 meV) and detected by a synchroscan streak camera with an overall time resolution of 20 ps. For decay times longer than 1 ns, the PL signal was analyzed by a time-correlated single-photon-counting system; in this case, the overall resolution was of the order of 200 ps.

The sample was held in a liquid-helium variable-temperature cryostat (4–300 K). The spot size on the sample was $240\times160\ \mu m^2$, so that 1 mW of average excitation power, in TRPL measurements, corresponds to a photogenerated carrier density of about $10^9\ cm^{-2}$. In the following, we will give, for simplicity, the average power

incident onto the sample; the corresponding carrier density can immediately be obtained from the previous equivalence.

III. RESULTS AND DISCUSSION

A. Influence of exciton localization on the PL decay time

Figure 1 shows the PL and PLE spectra of the 120-Å [Fig. 1(a)] and 180-Å [Fig. 1(b)] QW's at 4 K. A zero Stokes shift is observed between the PL line and the heavy-hole (HH) exciton line in the PLE spectrum. The full widths at half maximum (FWHM) are of the order of 0.8 and 0.6 meV, respectively, our spectral resolution being about 0.2 meV. The spectra corresponding to the other QW's are very similar; the FWHM increases for decreasing well width and a Stokes shift at most of the order of half the FWHM (HWHM) is found for the thinner wells (see Table I).

The dependence of the PL linewidth on the QW thickness L_w (Fig. 2) can be expressed at low temperatures as

$$\Gamma=\Gamma_{\text{in}}+\Gamma_0\ ,\qquad \Gamma_{\text{in}}=\delta L_w\left[\frac{\partial(E_e+E_{\text{hh}})}{\partial L_w}\right]\ , \tag{1}$$

where Γ_0 is the homogeneous linewidth of the PL line, mainly due to acoustical-phonon interaction and spin-flip scattering,[17] and Γ_{in} is the inhomogeneous spectral broadening originated by interface roughness (see Ref. 9, for example).

We find that the expression in Eq. (1) accounts satisfactorily for the experimental linewidths in Fig. 2 if one takes $\Gamma_0=160$ μeV and $\delta L_w=0.6$ Å; it should also be noted that the value of Γ_0 is in good agreement with the homogeneous linewidth of excitonic transitions in GaAs QW's, as obtained by time-resolved degenerate-four-wave-mixing (DFWM) experiments.[18] To summarize, we deal with a high-quality sample with low interface roughness, so that free-exciton properties are expected to be dominant. Moreover, in all five QW's, the integrated PL intensity is constant in the whole range of temperatures investigated, indicating that nonradiative processes play a negligible role, and that the measurements reported in the following refer mainly to radiative recombination.[8]

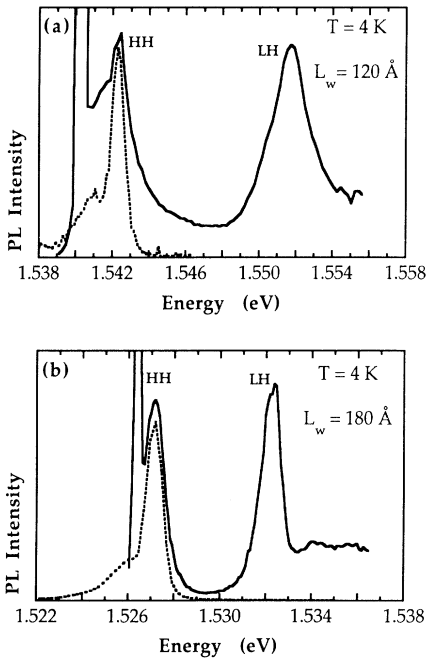

FIG. 1. PL (dotted line) and PLE (continuous line) spectra at 4 K: (a) 120-Å QW; (b) 180-Å QW.

TABLE I. Characteristic parameters of the quantum wells obtained from PL and PLE spectra at 4 K.

L_w (Å)	E_{PL} (eV)	Stokes shift (meV)	$\Delta E_{\text{LH-HH}}$ (meV)	FWHM (meV)
40	1.647	3.0	24.5	4.7
60	1.591	1.5	19.8	3.1
80	1.564	0.8	15.1	1.7
120	1.542	0	10.0	0.8
180	1.527	0	5.5	0.6

Figure 3(a) shows typical decay curves of the PL signal from the 120-Å QW at 14 K for three different excitation energies around the light-hole (LH) transition. We clearly observe an exponential decay of the PL signal in all cases and, if we assume a simplified three-level model, we can approximate the PL time behavior as given by the difference of two exponentials:

$$I_{\text{PL}}=I_0\frac{\tau_d}{\tau_r-\tau_d}[e^{-t/\tau_r}-e^{-t/\tau_d}]\ , \tag{2}$$

where τ_r and τ_d represent the rise time and the decay time, respectively, of the PL intensity. We will limit the discussion to the decay time τ_d; the results obtained for τ_r will be the subject of a separate paper.

The best fits obtained with Eq. (2) reproduce fairly well the experimental curves, as shown in Fig. 3(a) (full lines). Contrary to what is usually assumed, three different decay times are obtained for the three excitation energies; the longest decay time (400 ps) is measured when exciting resonantly the LH transition, while the fastest one (240 ps) is found for an excitation below the LH transition; an

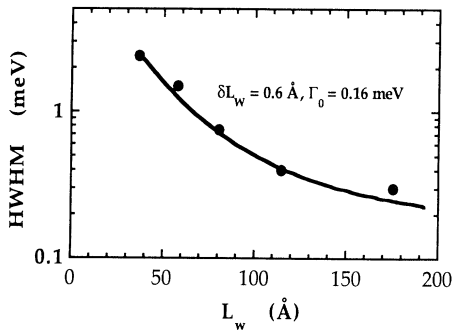

FIG. 2. The measured HWHM (full circles) at 4 K for all five QW's. The full line corresponds to a calculation using Eqs. (1) and (2), with $\delta L_w=0.6$ Å and $\Gamma_0=0.16$ meV.

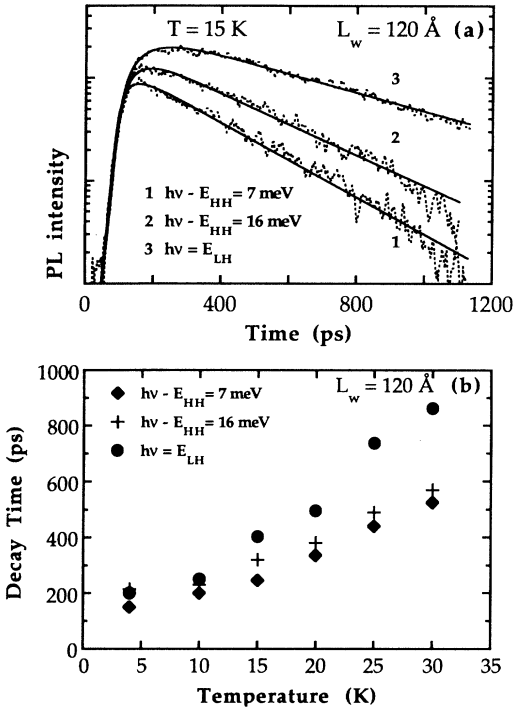

FIG. 3. (a) Experimental PL decay curves measured at 15 K after excitation (1) below, (2) above, and (3) at the LH transition, and the corresponding fits using Eq. (3). (b) The PL decay time of the 120-Å QW as a function of the temperature (4–30 K) for three different excitation energies: below (♦), above (+), and at the LH transition (●).

excitation above the LH transition gives an intermediate value (320 ps). Therefore, the problem arises of identifying a correct procedure for comparing the measured decay times with the exciton lifetimes predicted by the theoretical models. We believe that the origin of the different decay times depending on the excitation energy lies in the fact that luminescence from localized excitons is contributing to the measured decay time at the lowest temperatures.

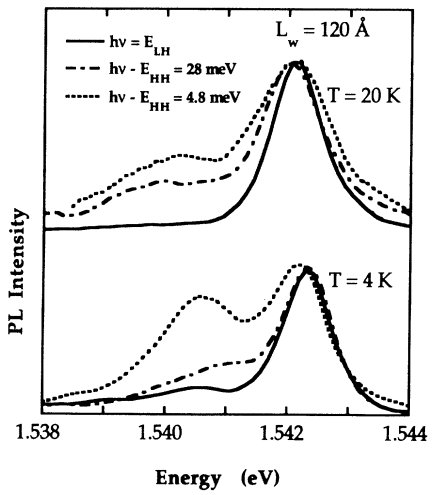

FIG. 4. The PL spectra of the 120-Å QW at 4 and 20 K for three different excitation energies: below (dotted line), above (dashed-dotted line), and at the LH transition (continuous line).

In Fig. 4, we report the cw PL spectra of the 120-Å QW at 4 and 20 K as a function of the excitation energy, $h\nu$. For this quantum well, the HH-LH exciton energy splitting is 10 meV (Table I), and the PL spectra have been recorded exciting the QW below ($h\nu - E_{HH} = 4.8$ meV), at ($h\nu = E_{LH}$), and above ($h\nu - E_{HH} = 28$ meV) the light-hole exciton transition. A secondary peak is observed 1.5 meV below the HH exciton transition, with an intensity depending on the excitation energy and almost disappearing when resonant excitation at E_{LH} is selected. At 20 K, the whole spectrum shifts down to lower energy according to the band-gap energy shrinkage, and the lower-energy peak becomes even less important.

We attribute the low-energy peak to partial localization of the generated free excitons (high-energy peak) at interface defects induced by local fluctuations in the confinement potential.[19,20] The relative intensities of the two peaks indicate that the luminescence is always dominated by recombination of free excitons, at least when exciting at or above the LH transition; partial migration to the lower-energy regions is only important when excitons are nonresonantly generated below the LH exciton transition, as indeed confirmed by the PLE spectra.

It follows that since typical spectral resolutions in TRPL measurements, as in our case, are about 1 meV, that is, about the energy separation of the two PL peaks, the measured decay time turns out to be an average over the characteristic recombination times of localized and free excitons, respectively, depending on the relative populations. The ratio between the two populations can roughly be estimated from the cw PL spectra; at 14 K, we find a ratio lower than 5% for LH excitation, 40% for $\Delta E = 7$ meV, and 25% for $\Delta E = 16$ meV. Consequently, the variation with temperature of the decay time also will be different for different excitation energies, as shown in Fig. 3(b).

We would like to conclude this section by stressing that mainly we generate free excitons only if we excite resonantly at the LH; in this case, the measured values and the increase of the PL decay time with temperature already below 10 K can safely be assigned to free-exciton features. For excitation below the LH transition ($h\nu - E_{LH} = -7$ meV), according to the more localized nature of the photogenerated excitons, the decay time does not increase appreciably up to 10–15 K, and it is significantly shorter than the free-exciton lifetime, in agreement with Ref. 21.

B. Temperature dependence of the free-exciton lifetime

From the previous discussion, we conclude that the identification of the PL decay time with the free-exciton lifetime at low temperatures can be made only for resonant excitation of the QW's as the excitonic transitions. Nonresonant excitation enhances exciton localization at potential fluctuations, and the observed decay time results are dependent on the relative populations of the free

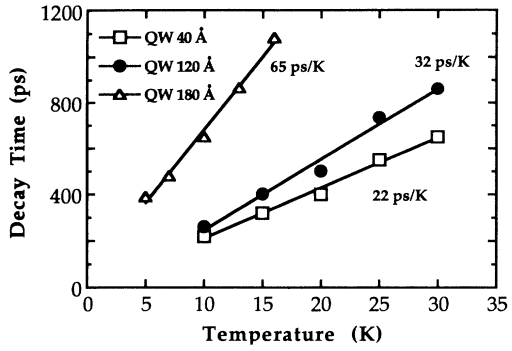

FIG. 5. PL decay times as a function of temperature (5–30 K) after excitation at the LH transition energy for the 40-Å (□), 120-Å (●), and 180-Å (△) QW's.

and localized excitons.

We expect, according to Andreani and co-workers,[4] a linear increase of the PL decay time for increasing temperature and resonant excitation of the QW's, with a slope depending on the exciton oscillator strength (or, equivalently, on the well width) that can be approximated, for $GaAs/Al_{0.3}Ga_{0.7}As$ QW's, by the simple expression[4]

$$\tau(T)=\frac{1793}{f_{xy}}T\ , \tag{3}$$

where τ is given in ps, the oscillator strength f_{xy} in units (10^{-5} Å^{-2}), T is the absolute temperature, the thermal average over the heavy-hole exciton band alone has been performed, and a common value of the photon wave vector k_0 has been assumed for all QW's.

In Fig. 5, we report the PL decay time of the 40-, 120-, and 180-Å QW's as a function of temperature in the interval 10–30 K; we do not consider the data below 10 K, in order to avoid possible contribution from localized excitons. A linear fit to the experimental points for all the QW's investigated yields the slopes reported in Table II; the oscillator strength values and the slopes predicted according to Eq. (3) are also reported in Table II. We see that the agreement between the theoretical and experimental slopes is indeed good except for the widest well (180 Å), where the experimental value is definitely larger than predicted. In fact, we expect the model by Andreani and co-workers[4] to underestimate the slope in wide wells, where the LH-HH exciton energy difference is so small that the LH band is already thermally populated at very low temperatures. Indeed, in the 180-Å well, we experimentally observe recombination at the LH transition even at 4 K, though it is very weak. If this is the case, given the smaller oscillator strength of the LH exciton compared to the HH exciton by a factor 3, from Eq. (3) we expect a value for the slope smaller than what would result from a thermal average over both the heavy and light exciton bands. We refer to Ref. 8 for a discussion of the exciton lifetimes at higher temperatures, where exciton ionization, non-negligible populations of the higher subbands, and free-carrier recombination, all outside the model by Andreani and co-workers,[4] have a major influence on the excitonic lifetimes.

TABLE II. Comparison of the experimental (τ^{exp}) and calculated (τ^{cal}) slopes for the temperature dependence of the free-exciton lifetime at low temperatures. τ^{cal} has been deduced, as explained in the text, from the values of the oscillator strengths f_{xy} deduced from Ref. 4 and reported in the second column. The errors on the best-fit values to the experimental data are around 10%.

L_w (Å)	f_{xy} (10^{-5} Å^{-2})	τ^{cal} (ps K^{-1})	τ^{exp} (ps K^{-1})
40	85	21	22
60	70	26	28
80	60	30	27
120	45	40	32
180	33	54	65

IV. CONCLUSION

We have performed cw and time-resolved PL measurements in good-quality single quantum wells at low temperatures. We have shown that resonant excitation at the LH transition produces a strong enhancement in the photogeneration of free excitons compared to excitons localized at interface fluctuations. As a consequence, resonant excitation makes it possible to study the lifetime of free excitons at low temperatures with negligible disturbance from localization effects. Indeed, after resonant excitation at the LH energy transition, a linear increase of the excitonic lifetime is observed in the interval 10–30 K, with a slope in agreement with the theoretical model for the radiative decay time of free excitons proposed by Andreani and co-workers.[4]

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