RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 46, NUMBER 11 15 SEPTEMBER 1992-I

Influence of barrier height on carrier lifetime in $Ga_{1-y}In_yP/(Al_xGa_{1-x})_{1-y}In_yP$ single quantum wells

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(Received 3 March 1992)

Using time-resolved photoluminescence, we have examined the photoluminescence (PL) decay time of $Ga_{1-y}In_yP/(Al_xGa_{1-x})_{1-y}In_yP$ single quantum wells with various well widths and different Al content in the barriers. At low temperatures, we find an increase of the lifetime with increasing temperature in good agreement with the temperature dependence of radiative recombination. At a characteristic temperature, which depends on the quantum-well thickness, a drop of the PL lifetime is observed. The temperature dependence can be explained by simultaneous thermal emission of electrons and holes out of the quantum wells. We find that the activation energy E_a is equal to one-half of the total confinement energy ΔE of the electron-hole pair in the quantum well and can be explained on the basis of Boltzmann statistics assuming thermal equilibrium between quantum-well and barrier states during the recombination.

The recombination of photoexcited carriers in quantum wells (QW's) has been extensively studied both experimentally[1–3] and theoretically[4–6]over the past few years. Despite a large number of studies there is no general agreement about the basic recombination mechanism, especially at high temperatures $T > 60$ K. On one hand, a series of papers[7,8] has attempted to establish the importance of the excitonic decay path up to room temperature, while some other authors[2,9] conclude that the photoluminescence (PL) lifetime at room temperature is controlled by nonradiative channels, and interface recombination gives the major contribution.

On the other hand, recent experiments by Bacher *et al.* have shown that carrier leakage into the barriers can influence the dynamic properties and the PL lifetime of shallow quantum wells.[10] Therefore, we cannot neglect the influence of carrier leakage into the barriers on the temperature dependence of the PL lifetime at higher temperature when carrier emission plays an important role. In this paper we present a detailed investigation of the temperature dependence of the PL lifetime in a set of $Ga_{1-y}In_yP/(Al_xGa_{1-x})_{1-y}In_yP$ QW's as a function of the well width L_z. The aim of the present paper is to show that the simultaneous thermal emission of carriers into the barriers leads to a strong reduction of the PL lifetime with an activation energy E_a equal to one-half of the total confinement energy ΔE in the quantum well.

The $Ga_{1-y}In_yP/(Al_xGa_{1-x})_{1-y}In_yP$ structures used in this study were grown by metal-organic vapor-phase epitaxy at a temperature of 710 °C lattice matched on GaAs substrates, which were oriented 6° off (100) toward ⟨110⟩. Each sample had a $Ga_{1-y}In_yP$ single quantum well with a width L_z varying in the range $1-10$ nm with emission energies between 1.984 and 2.100 eV, cladded by $(Al_xGa_{1-x})_{0.5}In_{0.5}P$ barriers [$E_{gap}(x = 0.3) = 2.144$ eV].

The measurements were performed using a standard photoluminescence setup, where the samples were excited by 5-ps pulses from a synchronously pumped mode-locked cavity-dumped dye laser. We used R6G, which made it possible to tune the excitation energy below the $(Al_xGa_{1-x})_{0.5}In_{0.5}P$ barriers. The luminescence was analyzed by a time-correlated single-photon counting technique providing a time resolution of the order of 150 ps.

A set of decay curves from the time-resolved PL measurements are shown in Fig. 1 for three different temperatures each measured at the peak of the emission line for a 3.5-nm well, together with the fits obtained by assuming a monoexponential decay. We find that the experimental data are consistent with pure exponential decay with time constant τ_{pl} over at least 2 orders of magnitude. This can be fully understood on the basis of the model outlined below. At low temperatures $T < 50$ K we also observe in all samples a weaker component with a

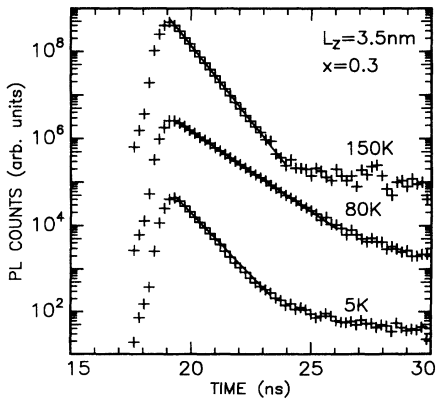

FIG. 1. Typical PL decay curves at three different temperatures for a 3.5-nm well. The solid curves give the fit to the experimental points assuming a monoexponential decay. The curves are vertically shifted and shown on a logarithmic scale.

longer decay time. This has also been observed in other materials,[3] but its origin is not clearly understood.

The temperature dependence of the PL decay times τ_{pl}, reported in Fig. 2, is similar for all the QW's investigated. At low temperatures ($T < 60$ K) we find an almost linear increase of the experimentally observed lifetime with increasing temperature which is in good agreement with the temperature dependence of radiative recombination in a two-dimensional semiconductor.[4,5]

At high temperatures a strong reduction of the lifetime is observed. The value of the temperature at which the maximum is reached depends on the quantum-well thickness. The activation energies (see Fig. 2) obtained from the measurements decrease with decreasing thickness of the quantum well. At the same time the quantization energy increases which leads to a smaller effective barrier confinement energy. If we compare the experimentally obtained activation energy E_a from Fig. 2 with the energetic difference between quantum-well and barrier luminescence at 2 K, we find that the thermal activation energy E_a is about one-half of the total confinement energy ΔE of the electron-hole pair in the quantum well (see inset of Fig. 2). This can be explained on the basis of a simultaneous thermal emission of electrons and holes in the barriers, which will be discussed in more detail below.

The interpretation of the strong reduction of the lifetime at high temperatures as a thermal activation is supported through the result shown in Fig. 3. There the PL lifetime is depicted versus temperature for two quantum wells with same well widths ($L_z = 3.5$ nm) but different Al contents x, which determine the confinement energy. In the sample with higher confinement energy ($x = 0.6$) the drop in the lifetime occurs at a higher temperature and with a higher activation energy.

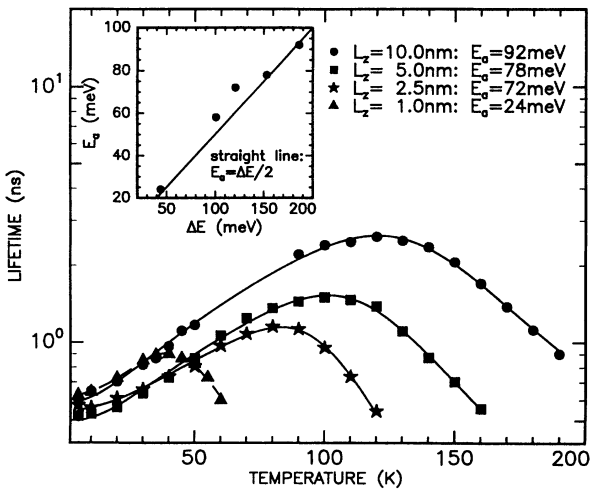

FIG. 2. Temperature dependence of the PL decay time τ_{pl} for different well thicknesses. The solid and the dashed lines represent fits as described in the text. The inset shows the activation energy E_a as a function of the confinement energy ΔE together with a straight line $E_a = \Delta E/2$.

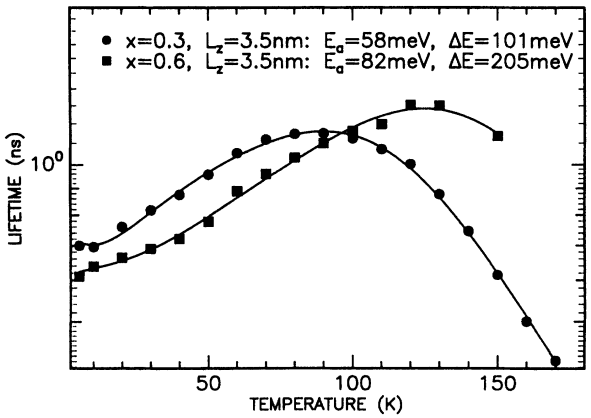

FIG. 3. PL lifetime vs temperature for different Al contents x in the barrier ($L_z = 3.5$ nm). The solid lines represent fits as described in the text.

In order to explain our experimental results for the temperature dependence of the lifetime, we make the following assumptions regarding the carrier kinetics. We assume that the carriers in the quantum well and in the barriers are in thermal equilibrium during the recombination process. This is a good approximation if the scattering time $1/C_{12}$ of carriers from the well into barrier states is much faster than the lifetimes τ_1 in the well and in the barriers τ_2 (see Fig 4). This assumption is in agreement with experiments performed by Bacher *et al.*[10] They have investigated carrier emission from $In_xGa_{1-x}As/GaAs$ QW's in the GaAs barrier with use of time-integrated and time-resolved spectroscopy. They found effective scattering times of about 10 – 100 fs between quantum-well states at the interface and barrier states.

Furthermore, we assume that the number of emitted electrons and holes in the barriers are equal ($\delta n_2 = \delta p_2$). Otherwise, if only one sort of carriers would be emitted into barrier states, they (e.g., electrons) would not have

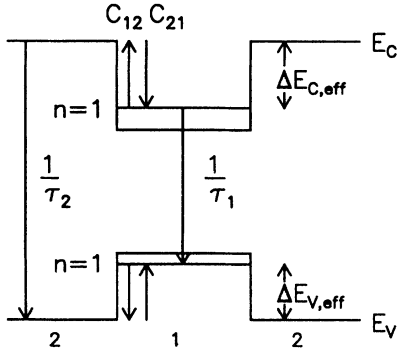

FIG. 4. Recombination of excess charge carriers in the quantum well $1/\tau_1$ and in the barriers $1/\tau_2$ and scattering C_{12}, C_{21} between quantum well and barrier states.

their corresponding partners (holes) in the barrier and could not recombine. This would lead to an electrical field, which would lower the barrier for carriers of opposite charge leading to an enhanced emission of such carriers until an equilibrium is reached.

Based on these assumptions we use the following system of rate equations describing the carrier dynamics in the low-injection regime:

$$\frac{d\delta n_1}{dt} = \frac{d\delta p_1}{dt} = -\frac{\delta n_1}{\tau_1} + C_{21}\delta n_2 - C_{12}\delta n_1, \tag{1}$$

$$\frac{d\delta n_2}{dt} = \frac{d\delta p_2}{dt} = -\frac{\delta n_2}{\tau_2} - C_{21}\delta n_2 + C_{12}\delta n_1. \tag{2}$$

Therefore, $\delta n_i, \delta p_i$ $(i = 1, 2)$ are the nonequilibrium densities of electrons and holes, τ_i are the carrier lifetimes, and C_{12}, C_{21} are the scattering rates between quantum-well states at the interface and barrier states.

From a detailed balance consideration,

$$\frac{d\delta n_1}{dt} = \frac{d\delta n_2}{dt} = 0, \tag{3}$$

we get under thermal equilibrium conditions

$$\frac{C_{21}}{C_{12}} = \frac{n_{1G}}{n_{2G}}, \tag{4}$$

where n_{iG} are the equilibrium carrier concentrations in the quantum well and barriers, respectively. In a first approximation, we consider parabolic bands and we ignore higher subband effects in the quantum well. In this approximation we get the relative concentrations δn_2 (δp_2) and δn_1 (δp_1) according to

$$\frac{\delta n_2}{\delta n_1}\frac{\delta p_2}{\delta p_1} = K_n K_p e^{-\Delta E_{C,\mathrm{eff}}/kT} e^{-\Delta E_{V,\mathrm{eff}}/kT} \tag{5}$$

with $K_i = (m_{i3d}^{3/2}/m_{i2d})(2\pi kT)^{1/2} L_z/h; i = (n, p)$. Therefore, $\Delta E_{C,\mathrm{eff}} = E_{C2} - E_{C1}$ $(\Delta E_{V,\mathrm{eff}})$ is the effective conduction (valence) -band discontinuity, m_{n3d} (m_{p3d}) and m_{n2d} (m_{p2d}) are the electron (hole) masses in the barrier and in the quantum well, and the quantities h, k are the usual fundamental constants.

With $\delta n_2 = \delta p_2$ and $\delta n_1 = \delta p_1$ we get

$$\frac{\delta n_2}{\delta n_1} = \sqrt{C} T^{1/2} e^{-\Delta E/2kT} = \phi. \tag{6}$$

Equation (6) describes the distribution of carriers between quantum-well and barrier states assuming thermal equilibrium between carriers in the quantum well and barriers. An important consequence of the assumption that the number of emitted electrons and holes in the barriers are equal is that we find a thermal activation energy equal to one-half of the total confinement energy $\Delta E = \Delta E_{C,\mathrm{eff}} + \Delta E_{V,\mathrm{eff}}$. This result is quite analogous to the well-known result for the equilibrium intrinsic carrier concentration $n_i \sim \exp(-E_g/2kT)$. As electrons and holes are thermally generated in pairs, their concentrations are equal, so that as a consequence only half the band-gap energy E_g determines the intrinsic carrier concentrations.

With the assumption that the carriers rapidly establish a dynamic equilibrium that persists throughout the recombination we can write

$$\frac{d\delta n_2}{dt} = \phi \frac{d\delta n_1}{dt}. \tag{7}$$

We can use this relationship in Eq. (1) to obtain a linear, first-order differential equation.

$$\frac{d\delta n_1}{dt} = -\frac{\frac{1}{\tau_1} + \frac{\phi}{\tau_2}}{1+\phi}\delta n_1. \tag{8}$$

Thus, solving Eq. (8) we obtain an effective lifetime $\tau_{\mathrm{eff}} = \tau_1(1+\phi)/[1 + (\tau_1/\tau_2)\phi]$ and with $\phi \ll 1$ which means that the carrier concentration in the quantum well is much larger than in the barriers, we finally get

$$\tau_{\mathrm{eff}} = \frac{\tau_1}{1 + \frac{\tau_1}{\tau_2}\sqrt{C} T^{1/2} e^{-\Delta E/2kT}} \tag{9}$$

which describes the temperature dependence of the lifetime observed in our experiments. At low temperatures, $(\tau_1/\tau_2)\sqrt{C}T^{1/2}\exp(-\Delta E/2kT) \simeq 0$ and therefore the effective lifetime τ_{eff} is controlled by the temperature-dependent lifetime τ_1 in the well. At higher temperatures we expect, from Eq. (9), $\tau_{\mathrm{eff}} \simeq \tau_2 \exp(\Delta E/2kT)/(\sqrt{C}T^{1/2})$ which leads to a strong reduction of the lifetime with increasing temperature, in agreement with our results.

The fit parameters we used are responsible for different parts of the lifetime curve. The activation energy E_a determines the thermal activated lifetime reduction at high temperatures, while the temperature at which the maximum of the lifetime is reached is mainly fixed by the fit parameter $(\tau_1/\tau_2)\sqrt{C}$. Taking for τ_1 the expression for the radiative excitonic lifetime derived by Feldmann, Peter, and Göbel,[4] we are able to fit our experiments using Eq. (9). The fits are shown in Fig. 2 and we find activation energies E_a of about one-half of the total confinement energy ΔE of the exciton in the quantum well in coincidence with our discussed model (see inset Fig. 2). Taking for C the known mass[11] and well thickness values, we obtain from the fits lifetime relations τ_1/τ_2 between 100 and 800. These large values are presumably due to the presence of nonradiative decay channels that shorten the lifetime in the barrier. We would like to emphasize that it is not excitons that are thermally emitted into the barrier, since the activation energies E_a, derived from our experiments, are much larger than typical exciton binding energies ($E_x < 12$ meV) in the quantum wells. Furthermore, we would like to point out that, following our analysis, one expects an activation energy equal to one-half of the total confinement energy if the observed intensity is proportional to the density of excess carriers in the wells. This is true for excitons as well as for the low injection case. It is interesting to consider the results of Bacher *et al.*,[10] where an activation energy equal to the total confinement energy was observed. Our model implies that these measurements were performed under high injection conditions, where $I \sim n^2$ holds.

It is very interesting to compare the recently published

PL lifetime measurements from Gurioli, Vinattieri, and Colocci[2] in $GaAs/Al_xGa_{1-x}As$ quantum wells with our results. They have also found a drop of the PL lifetime at high temperatures. It is also important noting that, for narrow wells, the maximum value of τ_{pl} is reached at lower temperature than in wider wells. Furthermore, the activation energy increases with increasing effective barrier height. They concluded that surface recombination and/or Shockley-Read recombination via states at the $GaAs/Al_xGa_{1-x}As$ interfaces give the main nonradiative decay channel. Based on our results we suggest an alternative explanation for their measured reduction of the lifetime, namely, thermal emission of carriers out of the quantum well.

In summary, we have studied theoretically and experimentally the dependence of the carrier lifetime in quantum wells on quantum-well thickness and temperature. We have demonstrated the importance of thermal emission of carriers out of $Ga_{1-y}In_yP/(Al_xGa_{1-x})_{0.5}In_{0.5}P$ quantum wells on electron-hole pair lifetimes. Our experiments show a thermal activation energy equal to one-half of the total confinement energy ΔE. This can be explained by the model presented which is based on the requirement of electrical neutrality, i.e., that the number of emitted electrons and holes in the barriers are equal.

We thank M. H. Pilkuhn for stimulating discussions. The financial support of this work by the Deutsche Forschungsgemeinschaft under Contract No. Ha 1670/1 is gratefully acknowledged.

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