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Influence of barrier height on carrier dynamics in strained $In_xGa_{1-x}As/GaAs$ quantum wells

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Strained $In_xGa_{1-x}As/GaAs$ heterostructures with different In content x, each containing quantum wells with various well widths L_z (between 2 and 30 nm) are investigated with use of time-integrated and time-resolved spectroscopy. With increasing temperature, a characteristic drop of both the photoluminescence (PL) intensity and PL lifetime is observed, revealing a strong dependence on In content and well width. The temperature dependencies can be explained by thermal emission of the carriers out of rather shallow quantum wells. The transfer of the carriers between quantum wells of different widths across the GaAs barrier is demonstrated and described by a system of rate equations.

Much interest is presently focused on lattice-mismatched heterostructures, which can be grown with excellent quality[1] and which are used in a variety of semiconductor devices. An important limitation for strained quantum wells is the existence of a critical thickness,[2] which basically correlates the energetic depths and the geometrical widths of the wells. Therefore, pseudomorphic $In_xGa_{1-x}As/GaAs$ quantum wells are usually characterized by a small confinement energy.

It is well known from other materials that carrier leakage into the barrier can influence the dynamic properties of quantum wells.[3,4] Therefore, we expect a modification of the carrier dynamics in shallow $In_xGa_{1-x}As/GaAs$ quantum wells. Recent experiments have shown a reduced carrier collection at high excitation densities and high temperatures[5] as well as reduced exciton scattering due to the low barriers[6] in strained $In_xGa_{1-x}As/GaAs$ quantum wells. We present in this paper a systematic study of the temperature dependence of the photoluminescence (PL) intensity and the PL lifetime in pseudomorphic $In_xGa_{1-x}As/GaAs$ quantum wells as a function of the In content x and the well width L_z. Thermal emission of the carriers out of the quantum well into the GaAs barrier[7] reduces both the PL intensity and PL lifetime in the quantum well. The carriers emitted by a narrow quantum well are transferred across the barrier and can be captured by a quantum well with larger well width L_z. The time-integrated and time-resolved experiments can be described quantitatively by using a system of rate equations.

The samples investigated consist of several $In_xGa_{1-x}As$ layers cladded by 200-nm GaAs barriers to avoid tunneling between the wells. Each sample has a different In content ($x=0.06$, 0.12, or 0.2) and contains four elastically strained quantum wells with $L_z=2$, 5, 10, and 30 nm (except for $x=0.2$, only $L_z=2$, 5, and 10 nm). Details of the sample characteristics and the growth procedure are given in Ref. 1.

For the experiments a cw-mode-locked Ar^+ laser followed by a synchronously pumped dye laser (pulse width 10 ps, $\lambda=650$ nm) with a repetition rate of 75.3 MHz was used. The PL signal was detected by a fast $S1$ microchannel plate (MCP). The experimental setup had an overall time resolution of about 50 ps. Low excitation powers ($P_{\text{peak}}=4$ kW/cm^2) were used to obtain excitonic emission. Figure 1 shows time-integrated low-temperature ($T=2$ K) PL spectra of all samples investigated. Sharp and intensive peaks due to free-excitonic emission indicate the high quality of the samples. Linewidths down to 0.35 meV are obtained in high-resolution measurements[1,6] and give evidence of an excellent sample quality.[6]

The dotted arrow in Fig. 1 marks the position of the barrier luminescence (1.515 eV), which can be detected at higher excitation power. As can be seen by the energy

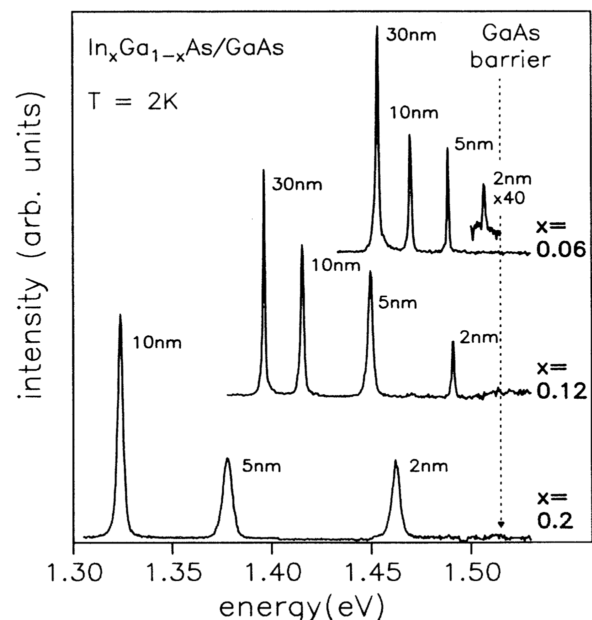

FIG. 1. Low-temperature ($T=2$ K) PL spectra of In_x-$Ga_{1-x}As/GaAs$ quantum wells with different In context x.

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difference of the quantum well PL and the barrier energy (dotted arrow), the samples are characterized by a small confinement energy ΔE, especially for those with low In content and small L_z. Even at $T=2$ K, the luminescence intensity of the smallest quantum well ($L_z=2$ nm) in the $In_{0.06}Ga_{0.94}As/GaAs$ sample, characterized by an extremely small confinement energy, is strongly reduced.

For a detailed study, we first investigated the temperature dependence of the PL intensity in the quantum wells. In Fig. 2 the PL intensity of an $In_{0.06}Ga_{0.94}As/GaAs$ sample is depicted as a function of temperature for different L_z using an Arrhenius plot [$\ln(I)$ versus T^{-1}]. At a characteristic temperature, which depends on L_z, a distinct drop of the PL intensity is observed. The intensity is reduced by more than 4 orders of magnitude in a small temperature range. At high temperatures, the slope of $\ln(I)$ versus T^{-1} is given by a characteristic activation energy E_A. If we compare the experimentally obtained activation energy E_A (from Fig. 2) with the difference between quantum well and barrier luminescence (Fig. 1), we find an excellent agreement. The thermal activation is therefore determined by the total confinement energy ΔE of the excitons in the quantum wells. A similar relation between the confinement energy ΔE and the thermal activation into the barrier is observed in all the samples investigated.

Based on this result, we use the following equation to describe the exciton dynamics:

$$\frac{\partial c}{\partial t}=-\frac{c}{\tau}-\frac{ce^{-E_A/kT}}{\tau_0}+g\,, \qquad (1)$$

where c is exciton concentration, τ is excitonic lifetime in the quantum well, and g is generation rate by the laser excitation. Due to the thermal distribution of the carriers in the quantum well, a certain part of the excitons [$c\exp(-E_A/kT)$] is scattered into the GaAs barrier, reducing the PL intensity of the quantum well. τ_0 is used as a fit parameter and describes the effective scattering time from the high-energy tail of the exciton distribution in the quantum well into the barrier states.

For steady-state conditions ($\partial c/\partial t=0$; $g=c_0/\tau$), Eq. (1) can be solved and we obtain

$$c(T)=\frac{c_0}{1+(\tau/\tau_0)e^{-E_A/kT}}\,. \qquad (2)$$

In a first approximation, the temperature dependence of τ/τ_0 is neglected with respect to the strong temperature dependence of $\exp(-E_A/kT)$. The solid line in Fig. 2 gives the theoretical fit, in good agreement with the experiment. A distinct decrease of intensity is already observed at temperatures $T\ll\Delta E/k$. The fit yields scattering times τ_0 between 10 and 100 fs, using exciton lifetimes of up to several nanoseconds. This result is in agreement with experiments performed by Oberli *et al.*[5] They have investigated the carrier capture from the GaAs barrier into an $In_{0.2}Ga_{0.8}As$ quantum well and found capture times shorter than their time resolution of about 1 ps, indicating a very fast scattering between barrier states and quantum well states at the interface.

A part of the thermally emitted carriers from a thin quantum well will be captured by the neighboring quantum wells with larger well width. To demonstrate this effect, we have plotted in the inset of Fig. 2 the temperature dependence of the PL intensity of the sample with $x=0.2$ only for the thinnest ($L_z=2$ nm) and the widest ($L_z=10$ nm) quantum well. As the intensity of the quantum well with $L_z=2$ nm drops due to the thermal activation of the excitons, we notice a distinct increase of the PL intensity in the larger quantum well. This clearly indicates carrier transfer between the narrow and the wider quantum well.

To obtain more detailed information on the dynamics of the carrier emission into the barrier, we performed time-resolved experiments using picosecond spectroscopy. In Fig. 3 the PL lifetime is depicted versus temperature for different well widths L_z ($x=0.2$; top) and different In

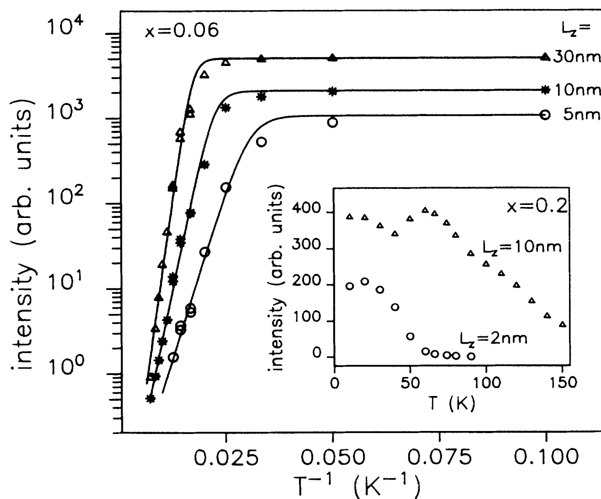

FIG. 2. Time-integrated PL intensity in $In_{0.06}Ga_{0.94}As/GaAs$ quantum wells depicted for different L_z in an Arrhenius plot (symbols: experiment; solid line: fit using Boltzmann statistics). The inset shows the temperature dependence of the PL intensity for the thinnest ($L_z=2$ nm) and the widest ($L_z=10$ nm) quantum well in a sample with an In content $x=0.2$.

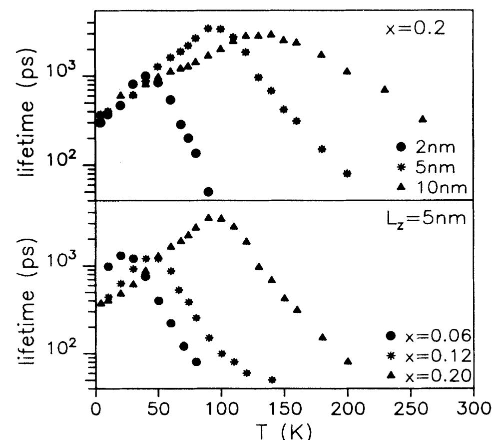

FIG. 3. PL lifetime vs temperature for different well widths L_z ($x=0.2$; top) and different In contents x ($L_z=5$ nm; bottom), respectively.

contents x (L_z =5 nm; bottom), respectively. At low temperatures, we find an increase of the lifetime with increasing temperature (up to around 3 ns), in qualitative agreement with the temperature dependence of excitonic recombination.[8] At a characteristic temperature, which depends on x and L_z, a strong reduction of the lifetime is observed. This drop occurs at higher temperatures if L_z or x, which determine the confinement energy, increases. Both the reduction of the PL intensity (Fig. 2) and the decrease of the PL lifetime (Fig. 3) are obviously caused by the same process, namely thermal activation of the excitons into the barrier.

For a theoretical description, we use a system of rate equations. In a first approximation, we consider two neighboring $In_xGa_{1-x}As$ quantum wells with different L_z, separated by a GaAs barrier. The exciton concentration in the thinner quantum well is given by c_1, whereas c_2 describes the concentration in the wider quantum well. If we assume a δ-function-like laser pulse, we obtain

$$\frac{\partial c_1}{\partial t}=-\frac{c_1}{\tau_1}-\frac{c_1 e^{-E_{A1}/kT}}{\tau_{01}}, \qquad (3)$$

$$\frac{\partial c_2}{\partial t}=-\frac{c_2}{\tau_2}-\frac{c_2 e^{-E_{A2}/kT}}{\tau_{02}}+\alpha\frac{c_1 e^{-E_{A1}/kT}}{\tau_{01}}. \qquad (4)$$

The excitonic lifetime in each quantum well is described by τ_i. ΔE_{Ai} and τ_{0i} are the activation energies and the effective scattering times from the quantum well into the barrier states, respectively.

The carrier concentration in each quantum well is reduced by both excitonic lifetime in the quantum well and exciton transfer out of the quantum well into the barrier. A part α of the thermally excited carriers of the smaller quantum well will be captured by the thicker one. The carrier capture and the relaxation into the ground state of the quantum well as well as the finite transfer time between the quantum wells across the barrier is very fast and can be neglected on the time scale of our experiment.[5]

Let us first focus on the solution of Eq. (3). With the initial condition $c_1(t=0)=c_{10}$, we get

$$c_1(t)=c_{10}e^{-t/\tau_{\mathrm{eff1}}}, \qquad (5)$$

with

$$\tau_{\mathrm{eff1}}=\frac{\tau_1}{1+(\tau_1/\tau_{01})e^{-E_{A1}/kT}}. \qquad (6)$$

Equation (6) describes the temperature dependence of the lifetime observed in our experiments. At low temperatures, $(\tau_1/\tau_{01})\exp(-E_{A1}/kT)\simeq 0$ and therefore the effective lifetime τ_{eff1} is controlled by the temperature-dependent radiative excitonic lifetime τ_1. At higher temperatures, we expect, from Eq. (6), $\tau_{\mathrm{eff1}}\simeq\tau_{01}\exp(E_{A1}/kT)$. This results in a strong reduction of the lifetime with increasing temperature, in agreement with our experimental results (see Fig. 3).

It is obvious from Eq. (4), that carrier transfer should change not only the transient behavior of the carrier concentration in the narrow quantum well but also that in the wider one. In the temperature region, where thermal emission dominates the excitonic lifetime in the narrow quantum well, the carrier concentration in the wider well $c_2(t)$ is approximately given by

$$c_2(t)=c_{20}e^{-t/\tau_{\mathrm{eff2}}}+\alpha c_{10}(e^{-t/\tau_{\mathrm{eff2}}}-e^{-t/\tau_{\mathrm{eff1}}}), \qquad (7)$$

with

$$\tau_{\mathrm{eff2}}=\frac{\tau_2}{1+(\tau_2/\tau_{02})e^{-E_{A2}/kT}}. \qquad (8)$$

τ_{eff1} and τ_{eff2} are the lifetimes in the narrow and the wide quantum well, respectively. The first term of Eq. (7) describes the temporal behavior of the carriers directly captured by the quantum well after the picosecond excitation. The transfer of the carriers into the wider quantum well is considered by the second part of Eq. (7). Therefore, the effective lifetime τ_{eff1} in the narrow quantum well influences the onset of $c_2(t)$ due to transfer.

In Fig. 4, we have plotted the time dependence of the emission intensity of the widest quantum well (L_z =10 nm) of an $In_{0.2}Ga_{0.8}As$/GaAs sample for different temperatures. The decay of the luminescence at delay times larger than 1 ns is determined by the effective lifetime, given by excitonic recombination at low temperatures and additional thermal emission of the carriers into the barrier at $T>150$ K. The onset of the emission is controlled by both, direct capture of carriers into the quantum well and transfer between the quantum wells.

In order to demonstrate the importance of the interwell transfer, we have fitted our experiments with two different models, one including (solid line) and the other neglecting (dotted line) carrier transfer between the quantum wells.

At low temperatures ($T\leq 50$ K; top trace in Fig. 4), no thermal emission of the excitons occurs. The lifetime of the 10-nm quantum well is given by excitonic recombination. No transfer between the quantum wells is expected, and both fits give identical results.

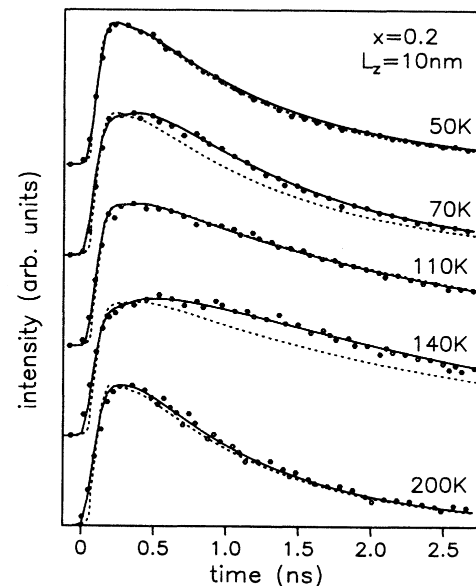

FIG. 4. Transient behavior of the PL intensity in an $In_{0.2}Ga_{0.8}As$/GaAs quantum well with L_z =10 nm for different temperatures (symbols). The solid and the dashed lines represent theoretical fits as described in the text.

At $T = 70$ K (second trace from top in Fig. 4), the lifetime in the narrowest quantum well ($L_z = 2$ nm) is strongly reduced due to carrier emission into the GaAs barrier (see Fig. 3). Taking for $\tau_{\mathrm{eff}1}$ the experimentally determined value of 285 ps (see Fig. 3), we are able to fit our experiments using Eq. (7), with $\alpha \simeq 0.6$ (c_{10} and c_{20} are obtained by the PL intensity at 2 K). This is a reasonable value for the capture probability, because a part of the thermally emitted carriers can reach the surface or the substrate or may be captured by the quantum well with $L_z = 5$ nm.

Although the transfer is most effective at higher temperatures, no transport effect is observed at $T = 110$ K (see the $T = 110$ K trace in Fig. 4). For these temperatures, the effective lifetime $\tau_{\mathrm{eff}1}$ in the 2-nm quantum well is very small (see Fig. 3). Therefore, we cannot resolve transfer-related effects in $c_2(t)$ with our time resolution.

If we raise the bath temperature further to 140 K, however, we observe again a significant influence of the interwell transfer (second trace from bottom in Fig. 4). In this temperature range, the excitons in the quantum well with $L_z = 5$ nm are thermally emitted into the barrier. If the temperature is increased further, thermal activation in the 5-nm quantum well results in a very small $\tau_{\mathrm{eff}1}$ and the transient behavior of $c_2(t)$ is once more unaffected by the transfer (bottom trace in Fig. 4).

In summary, we have presented time-resolved and time-integrated experiments demonstrating the importance of thermal emission of excitons out of $In_xGa_{1-x}As$ quantum wells with small confinement energy for the emission intensity and electron-hole pair lifetimes. We would like to emphasize that the present results have generally to be taken into account in a discussion of carrier dynamics in heterostructures with low barriers (e.g., $GaAs/Al_xGa_{1-x}As$ with low Al content or In_xGa_{1-x}-$As/In_yGa_{1-y}As_zP_{1-z}/InP$) and in thin quantum wells with only a few monolayer thickness, respectively. The effects studied here might be particularly important for the development of quantum-well lasers, in which a strong reduction of the quantum efficiency can occur at room-temperature operation.

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