Effect of indirect Γ-L and Γ-X transfer on the carrier dynamics of InGaP/InAlP multiple quantum wells

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Indirect Γ-L scattering within the well, and real space carrier transfer to the barrier X_{1c} states are shown to significantly affect the carrier dynamics in $In_{0.48}Ga_{0.52}P/In_{0.5}Al_{0.5}P$ multiple quantum wells. When carriers transfer to the indirect states occurs, the carrier dynamics is modified by the slow return of the carriers from the low mobility states to the well. As a result, the absorption recovery time increases by almost an order of magnitude. Carrier transfer to the indirect states also increases the carrier lifetime to values characteristic of indirect recombination. © *1997 American Institute of Physics.* [S0003-6951(97)03001-5]

$In_{0.48}Ga_{0.52}P/(Al_xGa_{1-x})_{0.5}In_{0.5}P$ multiple quantum wells (MQWs) are commonly used in the design of visible semiconductor lasers operating around 650 nm.[1] In the last few years, an effort in the design of visible laser diodes has been directed towards obtaining devices operating at wavelengths as short as 630 nm.[2] However, these devices have been found to have higher threshold current and higher temperature sensitivity than those operating at longer wavelength, characteristics which have been associated with poor electron confinement.[2]

Several factors can limit the electron confinement, such as a small conduction band discontinuity, or carrier transfer to indirect valleys. In the visible heterostructures the former can be essentially ruled out, as the conduction band discontinuity in the $In_{0.48}Ga_{0.52}P/(Al_xGa_{1-x})_{0.5}In_{0.5}P$ system has been measured to be 70% of the direct band gap difference between the well and barrier materials for a wide range of Al compositions x.[3,4] Scattering to the indirect valleys on the other hand, can significantly affect the carrier confinement, providing a leakage channel for carriers to transfer to the high effective mass L_{1c} or X_{1c} valleys. In bulk unstrained $In_{0.48}Ga_{0.52}P$, L_{1c} is approximately 0.2 eV below X_{1c} and can be accessible from Γ_{1c} by injecting carriers with excess energies of ~0.1 eV, the Γ_{1c}-L_{1c} separation.[5] In MQWs even lower energies are sufficient as the separation of the MQW ground state (Γ_{1e}) and L_{1c} is further reduced with increased carrier confinement. In addition, real space carrier transfer to the indirect X_{1c} valleys in the barrier (X^b_{1c}) may also occur in the $In_{0.52}Ga_{0.48}P/(Al_xGa_{1-x})_{0.5}In_{0.5}P$ MQWs when the barrier composition x is selected to be larger than 0.5 and the barrier becomes indirect. The associated decrease in the carrier mobility that accompanies this transfer has been shown to have dramatic effects on the threshold current of $In_{0.48}Ga_{0.52}P/(Al_xGa_{1-x})_{0.5}In_{0.5}P$ lasers.[6]

In this letter we present evidence of the important of Γ-L scattering and real space Γ-X carrier transfer on the carrier dynamics of $In_{0.48}Ga_{0.5}P/In_{0.5}Al_{0.5}P$ MQWs and show that when carrier transfer to indirect states occurs, the carrier dynamics becomes dominated by the slow return of the carriers from the low mobility states to the well. As a result, the absorption recovery time increases by an order of magnitude. The carrier lifetime is also increased reaching values characteristic of indirect recombination.

The effect of carrier transfer to indirect states in $In_{0.48}Ga_{0.52}P/In_{0.5}Al_{0.5}P$ MQWs was evaluated from time resolved absorption (TRA) measurements as a function of excitation wavelength and independently from time resolved photoluminescence (TRPL) measurements at high pressure. The TRA and TRPL experiments are conceptually different; however, they provided an independent assessment of the indirect carrier transfer. In the TRA experiments, carrier transfer to L_{1c} in the well and to the barrier X^b_{1c} states was achieved by selectively photopumping carriers into the well with sufficient excess energies to reach the indirect states. The dynamics of the photoexcited carrier population was monitored from the absorption changes of a weak probe beam delayed with respect to the pump beam. In the TRPL the dynamics of carrier transfer was evaluated from the changes in the PL lifetime (τ_{PL}) as the separation of Γ_{1e} and the indirect L_{1c} and X^b_{1c} states was modified by using hydrostatic pressure.

The $In_{0.48}Ga_{0.52}P/In_{0.5}Al_{0.5}P$ MQWs used in these studies were grown by gas source molecular beam epitaxy on [100] GaAs and consisted of 30 periods of 85 Å wells separated by 225 Å barriers. The MQWs were nominally undoped ($N_d - N_a \leq 10^{16}$ cm^{-3}).[7] The TRA measurements were conducted using a pump and probe configuration, with both beams being generated by a synchronously pumped dye laser, which produced pulses of ~2 ps.[8,9] Two different dye combinations, Rhodamine 6G (R6G) and DCM, were used as the laser active medium to allow for wavelength selectivity in the range 580–600 nm (2.14–2.07 eV) and 600–650 nm (2.07–1.91 eV), respectively. The TRPL measurements at high pressure were conducted with the dye laser operating at 580 nm.[4] The excitation density at the sample was calculated to be 3×10^{11} cm^{-2}. The photoluminescence decay was measured using a Hamamatsu M1955 synchroscan streak camera with a resolution of 10 ps. The TRA and

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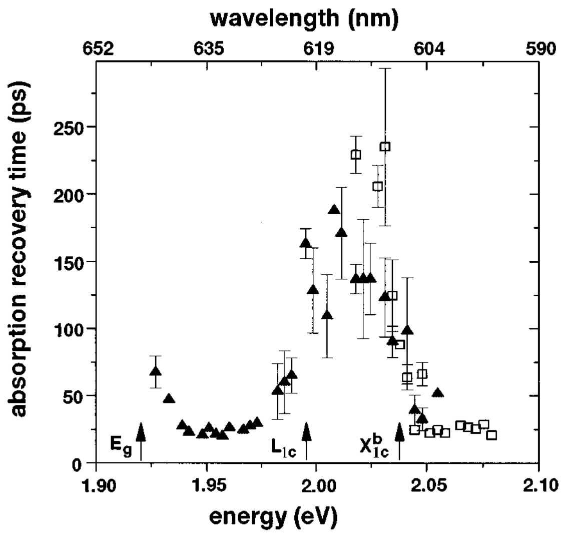

FIG. 1. (a) Variation of the absorption recovery time with excitation energy. ▲ and □ identify the decays obtained by pumping with R6G and DCM, respectively. The arrows mark the energy position of the band gap E_g, and that of L_{1c} and X^b_{1c} with respect to E_g.

TRPL experiments were both performed at room temperature.

The variation of the absorption recovery time (τ_a) with excitation energy is shown in Fig. 1. τ_a decreased from 75 ps to 20 ps for energies between 1.925 and 1.975 eV. Further increase of the photon energy dramatically increased τ_a which reached values as large as 200 ps for photon energies around 2.02 eV. This behavior can be understood in relation to the conduction band structure of the $In_{0.48}Ga_{0.52}P$/$In_{0.5}Al_{0.5}P$ MQWs,[4,5] schematically shown in Fig. 2. Around 1.92 eV, photoexcited carriers are created into Γ_{1e} with very small excess energies and τ_a is basically the recombination lifetime, determined from TRPL measurements to be 80 ps. For energies between 1.93 and 1.98 eV, $\tau_a = 20$ ps is a measure of the intraband relaxation time from the 30 meV wide optically coupled region. The increase of τ_a for energies between 1.98 eV and 2.02 eV can be explained by considering that in this energy range photoexcited carriers are created into Γ_{1e} with sufficient excess energy to transfer to L_{1c}. Although this transfer is typically less than 100 fs,[10] the return of the carriers from the low mobility states to the well is much slower and thus τ_a increases. For photon energies above 2.03 eV carriers are also created in Γ_{2e}, which is almost resonant with both L_{1c} and X^b_{1c}. Thus in addition to Γ-L scattering, a new transfer channel which involves real and k-space transfer is opened for the carriers. This carrier transfer mechanism has been found to be very efficient in GaAs/AlAs MQWs.[11]

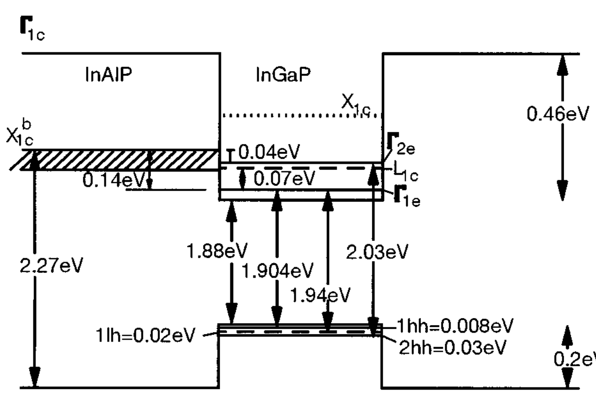

FIG. 2. Band structure of the 85 Å/225 Å $In_{0.48}Ga_{0.52}P/In_{0.5}Al_{0.5}P$ MQWs, showing the separation of the electronic conduction states associated with Γ_{1c}, L_{1c}, and X_{1c}. The shaded region corresponds to the energy range for which long recovery times are measured.

To estimate the effect of the intervalley Γ-L and real space Γ-X transfer processes on the carrier dynamics, we calculated the corresponding scattering rates. We assumed that L_{1c} and X^b_{1c} were quantized and calculated the rates assuming intervalley deformation potential scattering.[12] For this mechanism, the rate is dependent on the overlap of the square of the wavefunctions of the initial and final states, and thus should yield much larger values for the Γ-L scattering rate than for the Γ-X^b_{1c} transfer rate, for which the wavefunctions are spatially separated. The Γ_{1e}-L_{1c} and Γ_{2e}-L_{1c} scattering rates were calculated to be 6 ps^{-1} and 2 ps^{-1},[13] similar to intervalley scattering rates measured in bulk GaAs.[10] The rate of return of the carriers from L_{1c} to the well was calculated by multiplying the Γ-L scattering rate by the ratio of the density of state mass of L_{1c} and Γ_{1c}.

The Γ-X^b_{1c} transfer rate was also calculated using intervalley deformation potential scattering and by taking into account state mixing of the well Γ and barrier X^b_{1c} states close in energy.[14] The wavefunctions of the mixed initial and final state were expressed as a linear combination of the unperturbed Γ and X^b_{1c} wavefunctions. The mixing coefficient was taken equal to $A/\Delta E$, where A is a phenomenological coupling coefficient (~3 meV)[14] and ΔE is the Γ-X^b_{1c} energy difference. The Γ_{2e}-X^b_{1c} and Γ_{1e}-X^b_{1c} transfer rates were calculated to be equal to 0.3 ps^{-1} and 6×10^{-3} ps^{-1}, respectively.

Using the calculated Γ-L scattering and Γ-X transfer rates, and the measured intraband relaxation time, we modeled the evolution of the photoexcited carrier distribution in levels Γ_{1e}, Γ_{2e}, L_{1c} and X^b_{1c} using a 4 level system of rate equations.[15] The Γ_{1e} and Γ_{2e} carrier populations were allowed to transfer to L_{1c} and X^b_{1c} and to relax within the well with interband and intraband rates of 0.1 ps^{-1} and 0.05 ps^{-1}, respectively. For the L_{1c} and X^b_{1c} populations we only included transfer to Γ_{1e} and Γ_{2e} and did not allow L_{1c}-X^b_{1c} transfer, as this process should be very improbable due to the large change in momentum required. The simulations showed that when only Γ-L scattering is included, the return of the carriers into the well states increases the relaxation time to ~130 ps, as found experimentally. When the transfer channel to X^b_{1c} is opened, the slower dynamics of carrier transfer to and from X^b_{1c} increases the carrier decay to values of the order of 270 ps, in agreement with the experimental results.

The Γ-X transfer was independently evaluated from TRPL measurements. Γ_{1e}-L_{1c} transfer was not observed in these experiments, as it is masked by the Γ_{1e}-X^b_{1c} transition which occurs at a crossover pressure $P_c = 1.13$ GPa.[4] Below P_c, the PL spectra was that characteristic of Γ_{1e}, and had a lifetime $\tau_{PL} = 80$ ps. This short carrier decay indicates that τ_{PL} is dominated by nonradiative processes.[16] Very close to

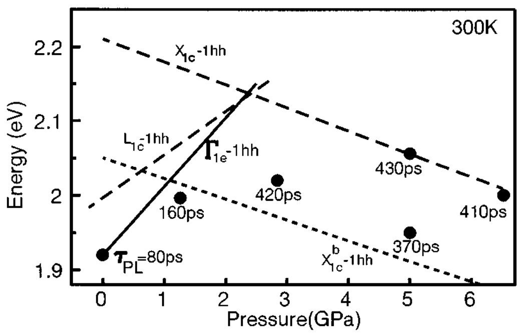

FIG. 3. Lifetime (τ_{PL}) and associated peak energy of PL transitions in $In_{0.48}Ga_{0.52}P/In_{0.5}Al_{0.5}P$ MQWs at high pressures. The straight lines show the pressure dependence of the PL transitions determined from cw PL measurements of Refs. 4 and 5.

P_c, τ_{PL} increased to 160 ps and at higher pressures τ_{PL} increased to 400 ps, a value characteristic of indirect recombination in these MQWs.[4] The measured τ_{PL} and their associated PL peak energy are plotted as a function of pressure in Fig. 3.

The variation of τ_{PL} with pressure can be explained by taking into account the effect of intervalley and interlayer Γ_{1e}-X^b_{1c} mixing that occurs when these states are close in energy.[17] Far away from P_c, the PL lifetimes τ_{PLI} and τ_{PLII} correspond to those of the unperturbed Γ_{1e}-1hh state below P_c and X^b_{1c}-1hh above P_c. Very close to P_c, when mixing occurs, the carrier lifetime of the staggered $X^b_{1c}-1$hh transition decreases below 400 ps, as its radiative component decreases with ΔE, the Γ_{1e}-X^b_{1c} separation. We calculated τ_{PLII} by considering the contribution of the radiative (τ_{IIr}) and nonradiative lifetimes (τ_{IInr}), as $\tau_{PLII}=(1/\tau_{IIr}+1/\tau_{IInr})^{-1}$. Γ-X mixing was taken into account by expressing $\tau_{IIr}\sim\tau_{1r}(\Delta E/A)^2$, where τ_{Ir} is the radiative lifetime of the $\Gamma_{1e}-1$hh transition.[18] We also assumed τ_{IInr} to be independent of pressure.[19] These relationships show that close to P_c, τ_{PLII} is basically determined by the variation of τ_{IIr} with pressure (or ΔE) and above P_c saturates at the value of τ_{IInr}. Using values of $A=3$ meV, $\Delta E=5$ meV at 1.2 GPa,[4] $\tau_{Ir}\approx 1.4$ ns[19] and $\tau_{IInr}=400$ ps, we calculated $\tau_{PLII}=360$ ps. The factor of 2 difference between the calculated and measured τ_{PLII} suggests that ΔE may be smaller than 5 meV. A lower ΔE is expected in this case, as the $X^b_{1c}-1$hh peak energy blueshifts with excitation power,[4] and the pump power is almost twice that used in Reference 4.

The results of our experiments have important consequences for the design of visible semiconductor lasers, as they show that carrier transfer to the indirect states within the well and to the barrier significantly affects the carrier dynamics. L_{1c} scattering is expected to become important in narrow lattice matched and in tensile strained InGaP/InAlP MQWs, in which the Γ_{1e}-L_{1c} separation is small.[5] Carrier transfer to X^b_{1c} is also expected to modify the output characteristics of MQW lasers, when the separation of the lowest confined states and X^b_{1c} is small, as it occurs in highly compressive and tensile InGaP/InAlP MQWs.[5]

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