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ULTRAFAST CARRIER CAPTURE IN QUANTUM WELL STRUCTURES

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Abstract

We present an experimental and theoretical study of the carrier capture time into a semiconductor quantum well. The carrier capture time was obtained by measuring both the rise of the quantum well (QW) population using time-resolved luminescence measurements and the decay of the barrier population using pump-probe correlation experiments. In the first technique, we compare the QW rise-times after direct (below the barrier band gap) and indirect (above the barrier band gap) excitation in order to eliminate the effects of relaxation and exciton formation in the quantum well.

We report the first experimental observation of oscillations in the carrier capture time between 3 and 20 ps as a function of quantum well thickness, obtained from both techniques. The observed capture times are, for the first time, in agreement with theoretical predictions from an ambipolar capture model.

Key Words: Quantum well, carrier capture, lasers, time resolved luminescence, LO phonons, oscillating capture time, carrier cooling time, resonant exciton absorption.

Introduction

The carrier capture time has first been calculated by Brum and Bastard (1986) who predicted an oscillating capture time between 30 ps and 1 ns as a function of quantum well (QW) thickness. The capture process, which is governed by the emission of LO phonons, is enhanced whenever a barrier state couples into the quantum well, giving rise to a large wavefunction overlap and thus to efficient capture. With respect to the carrier capture time, it is a major question whether the predicted oscillations in the carrier capture time do exist or not. In other words, is the quantum mechanical approach, which predicts these oscillations, correct or is a classical approach adequate?

Until recently, the predicted oscillations of the carrier capture time were not observed and most experimental results (Deveaud *et al.*, 1988; Kersting *et al.*, 1992; Weiss *et al.*, 1992) yielded much smaller capture times than predicted by Brum and Bastard. Only recently, oscillating capture times have been observed by three different groups (Blom *et al.*, 1993a; Morris *et al.*, 1993; Barros *et al.*, 1993) which are not only qualitatively but also quantitatively in agreement with the approach of Brum and Bastard. The major difference with the older work is that we measured the capture time in carefully designed separate confinement heterostructure single quantum wells (SCH-SQW) as shown in Figure 1, and that we carefully separated the capture time from other relaxation times in the barrier and inside the quantum well.

Theory of the Carrier Capture Time

After injection into the barrier layer of a SCH-SQW structure, the carriers relax very fast (< 1 ps) by LO phonon emission until they are within one LO phonon energy of the lowest barrier state. Then, the carriers are scattered from the lower barrier states into the quantum well. The dominant scattering mechanism for carrier capture at low carrier densities is LO phonon emission, by which the carriers lower their energy by 36.8 meV. For SCH-SQW structures in which the barrier width is of the same order of magnitude as the coherence length

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Figure 1. Schematic of the conduction band structure of the multiple SCH-SQW sample used in the experiments. The structures consist of 10 identical single quantum wells separated by 50 nm SCH barriers and 10 nm AlAs confinement layers at both sides. The carriers rapidly cool down inside the AlGaAs barrier layer until they are within 36 meV of the lowest barrier state. Carrier capture occurs by LO-phonon emission from a barrier level into the quantum well. Finally, the carriers will further cool down inside the quantum well. The energy levels in the barrier appear in pairs with wavefunctions of odd and even symmetry.

of the carriers, the capture process is expected to be dominated by quantum mechanical effects. We consider a GaAs/Al_xGa_{1-x}As SCH-SQW in which both the quantum well states and the barrier states are 2-dimensional. The transition probability $W_{k,k'}$ for a carrier in an initial barrier state with wave vector k to emit a LO phonon and to become captured into a bound state in the quantum well with wave vector k' is given by:

$$W_{k,k'} = (2\pi/\hbar) \{1/(2\pi)^3\} \int dq_z |I(q_z)|^2 |c|^2 \delta(k-k-q) |_{xy} \delta(E_i(k) - E_f(k') - \hbar\omega), \quad (1)$$

with q_z the phonon wave vector parallel to the growth axis and $|c|$ the Frohlich coupling constant for LO phonons. $I(q_z)$ is the overlap integral between the quantum well and the barrier envelope functions which shows strong resonances each time when a new bound level is coupled into the quantum well. $I(q_z)$ is given by:

$$I(q_z) = \int dz \exp(-iq_z z) \Psi_f^*(k') \Psi_i(k). \quad (2)$$

The total scattering rate for a transition from an initial state to the allowed final states is given by:

$$\tau(k)^{-1} = \int W_{k,k'} dk'. \quad (3)$$

The experimentally observed capture time is, of

Figure 2. Calculated dependence of the quantum mechanically calculated overall capture time on the SCH-barrier thickness (at one side) for a SCH-SQW structure with a 5 nm quantum well.

course, also dependent on the carrier distribution in the barrier layer, which is further explained in **Dependence on the Carrier Distribution in the Barrier**. Our calculated capture times will be presented later (in Figure 6, in which we have plotted the ambipolar capture time; Blom *et al.*, 1993a):

$$\tau_a = \{(\tau_e + \tau_h)/(2\tau_e\tau_h)\},$$

in which the electrons have been treated quantum mechanically and the holes have been described by a classical diffusion process. It is further assumed that no LO phonons are present, so there is no thermally activated escape from the quantum well which is correct at low temperatures.

Our quantum mechanically calculated capture time is valid for carriers in the whole separate confinement heterostructure (SCH) barrier and is, therefore, an overall capture time. The overall capture time is roughly proportional to the SCH-barrier thickness through the overlap integral $I(q_z)$, as shown in Figure 2. At SCH-barriers below 20 nm, the number of confinement levels in the barrier is too small for observing a smooth behavior, whereas computational instabilities are observed above 100 nm. The linear behavior we find in Figure 2 is in contrast to the classical "diffusion limited" capture time, which is proportional to the square of the SCH-barrier thickness.

We want to point out that the capture time is inversely proportional to the number of quantum wells since the capture time is inversely proportional to the number of final states as shown in eq. (3). Consequently, in measurements of the capture time in multi quantum well (MQW) structures (Deveaud, 1988; Kersting

Figure 3. Determination of the capture time from the decay of the barrier population in a SCH-SQW with a 7 nm quantum well. In the upper part, the decay of the barrier population is observed as an increase of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ probe luminescence as a function of pump-probe delay. In the lower part, the decay of the barrier population is observed as a decay of the probe luminescence of the GaAs buffer layer versus pump-probe delay showing an increasing probe transmission through the SCH-SQW structures versus time.

et al., 1992), the observed capture times were below 1 ps, which is full in agreement with theory. In these structures, an experiment measures the sum of a very small but oscillating capture time and a non-oscillating cooling time in the quantum well and in the barrier.

Observation of an Oscillating Capture Time

Decay of the barrier population

The capture times of photoexcited carriers in an undoped SCH-SQW structure are experimentally measured at low temperatures (8K) by two different techniques with picosecond resolution, measuring the rise-time of the quantum well population and the decay time of the barrier population.

Figure 4. Experimental determination of the carrier capture time using rise-time differences of the quantum well luminescence after direct (closed circles) excitation into the quantum well and indirect (open circles) excitation into the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers with a laser pulse (0.6 ps). A capture time of 19.1 ps is derived for this structure. The experiments (8K) were carried out at an excitation density of $2 \cdot 10^{16} \text{ cm}^{-3}$ in a SCH-SQW structure with a 2.6 nm quantum well.

In the latter technique, the decrease of the carrier concentration in the barrier states is measured. We have performed two-pulse correlation experiments on the luminescence of the barrier states. In a two-pulse correlation experiment, the PL-decay of the barrier layers shows up as an increase of the correlated PL-signal. This can be understood as follows: A strong excitation pulse ($2 \cdot 10^{17} \text{ cm}^{-3}$) creates electrons and holes in the barrier states and reduces the absorption of a second laser pulse ($1 \cdot 10^{17} \text{ cm}^{-3}$) with the same photon energy. If, however, the delay between the laser pulses exceeds the carrier capture time, then the absorption of the second laser pulse is no longer reduced. As a result, the barrier luminescence due to the probe pulse is expected to increase with increasing time delay between the laser pulses until the carriers of the first pulse are captured by the well. In Figure 3, the correlated barrier luminescence is shown for a 7 nm quantum well as a function of the delay between the laser pulses together with a fitted curve, which indicates a capture time of $(14.8 \pm 2) \text{ ps}$. The temporal width of the laser pulses was 0.6 ps and the excess energy of the pump and probe pulse (2.014 eV) with regard to the barrier band gap (1.981 eV) was 33 meV. The detection energy was set to the maximum of the barrier luminescence (1.987 eV) with a broad detection window of 10 meV.

The fact that the increase of the correlated barrier

Figure 5. Observation of an oscillating carrier capture time. The figure shows the excellent agreement between the experimentally observed capture times with the theoretical calculated ambipolar capture times. The experimental data have been obtained by (○) upconversion measurement of the difference in QW rise-times, by (Δ) two-pulse correlation measurements of the population decay in the barrier layers and by (■) "transmission-like" correlation experiments detecting the bulk GaAs PL-signal.

luminescence is actually due to an increase of the probe absorption is confirmed by a "transmission" type of experiment in which we use the bulk GaAs buffer layer which is located between the 10 SCH-SQW structures and the substrate as a kind of "photodetector" detecting the probe transmission through the SCH-SQW structures. Specifically, we detect the bulk luminescence intensity from the buffer layer which is proportional to the probe transmission. The probe transmission measures the amount of bandfilling in the AlGaAs layer of the SCH-SQW structure which is large directly after the pump pulse, but which decreases in time when the barrier population is emptied into the quantum well. Consequently, the probe transmission is expected to decrease, resulting in a decrease of this GaAs luminescence as a function of the pulse delay, as is shown in Figure 3b. As expected, the time constant of the increase of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ luminescence (Fig. 3a) is also found in the decrease of the GaAs luminescence (Fig. 3b) as a function of the delay between the laser pulses.

Rise-time of the quantum well luminescence

In this technique, we deduce the carrier capture times from luminescence rise-time measurements using an upconversion lightgate. The capture times are determined from the differences in the rise-time of the quantum well luminescence after direct (below the barrier band gap) and indirect (above the barrier band gap) excitation with a subpicosecond laser pulse (0.6 ps), in order to eliminate the effect of relaxation of the carriers in the quantum well. In Figure 4, the time evolution of the quantum well luminescence is shown after direct and indirect excitation. By fitting the experimental rise-times to the rate equations, we obtain a capture time of 19.1

± 2 ps for this well width. Furthermore, no dependence of the capture time on the excitation density was found in the range $3 \cdot 10^{15} \text{ cm}^{-3}$ to $2 \cdot 10^{17} \text{ cm}^{-3}$. For larger excitation densities, no capture times could be obtained due to band filling in the quantum well.

We measure the carrier capture time as the difference of the PL-rise-time after indirect and direct excitation where the excitation energy for direct excitation is precisely one LO-phonon energy (36 meV) below the energy for indirect excitation. Consequently, with direct excitation, we measure the cooling time of the carriers starting from the energy level where they end up after the capture process towards the exciton energy where the luminescence is detected. These precautions are necessary because the cooling time inside the quantum well also oscillates as a function of the excess carrier energy due to resonant exciton formation (Blom *et al.*, 1993b), i.e., the carrier cooling time is smaller when the excess energy of the carriers is precisely adequate to form an exciton at $k = 0$ after cooling down by emitting a phonon cascade.

Oscillating carrier capture time

We have measured the capture time for a set of SCH-SQW samples with different quantum well thickness. The variation of the observed carrier capture times as a function of well width is shown in Figure 5. We, for the first time, find oscillations in the carrier capture time between 3 and 20 ps as a function of quantum well width in a time-resolved experiment. It should be noted that for all samples, the capture times obtained from the correlation measurements reproduce within experimental error (± 2 ps) with the capture times derived from the quantum well rise-times. Also included are the

predictions of our ambipolar capture model (Blom *et al.*, 1993a) presented in **Theory of the Carrier Capture Time**. The observed capture times are in excellent agreement with the theoretically predicted ambipolar capture times without using any fitting parameters. Similar oscillations of the capture time versus quantum well thickness have recently also been reported by Barros *et al.* (1993) and by Morris *et al.* (1993).

We performed similar upconversion measurements, as presented in Figure 4, at $T = 77\text{K}$ in order to investigate the temperature dependence of the carrier capture time. A high enough increase of the temperature is expected to decrease the carrier capture efficiency due to an enhanced LO phonon absorption, which gives rise to an increase of the carrier escape from the well. From the rise-time difference, a capture time of $17.7 \pm 2\text{ ps}$ was obtained at 77K, which equals the capture time of $19.1 \pm 2\text{ ps}$ measured at $T = 8\text{K}$ within experimental error. Therefore, no temperature dependence of the carrier capture time is observed up to 77K.

Dependence on the Carrier Distribution in the Barrier

A final point to discuss is the dependence of the capture time on the carrier distribution in the SCH-barrier layer. The carriers will rapidly cool down by LO-phonon emission until their excess energy in the SCH-barrier is less than 36 meV. Such a rapid cooling is expected since the LO-phonon emission time is approximately 0.1 ps, which is much smaller than the capture time. For carrier capture, only the dependence of the capture time on the carrier distribution up to 36 meV above the SCH-barrier is relevant.

We investigated the dependence of the carrier capture times on the population of the barrier sub-bands after optical excitation at a low excitation density in a SCH-SQW structure with a quantum well width of 5 nm. At a laser excitation excess energy of 36 meV, as plotted in Figure 6, we observe a sharp decrease of the carrier capture time from 20 to 6 ps. The 5 nm quantum well contains two bound states for the electrons, located at 190 meV and 4 meV below the barrier band gap respectively. It should be noted that a laser excess energy of 36 meV with regard to the barrier band gap corresponds to an excess energy of 32 meV for the electrons and to an excess energy of only 4 meV for the heavy holes, due to the differences in the effective masses. Electrons with an excess energy larger than 32 meV are able to emit a 36 meV LO phonon and are captured into the second sub-band, which is located only 4 meV below the barrier band gap. The electrons, which have an excess energy of less than 32 meV with regard to the barrier band gap, are only able to make a

Figure 6. Calculated (solid line) and experimental (closed circles) capture times as a function of laser excess energy for a 5 nm quantum well structure. The sudden jump of the capture time around 32 meV proves that phonon emission is the dominant capture mechanism and also shows that the capture time is heavily dependent upon the barrier population.

transition to the lowest bound state. Thus, we show that the experimental capture times strongly depend on the population of the energy levels in the barrier.

Furthermore, the sharp decrease of the carrier capture time demonstrates the dominance of the LO phonon emission in the capture process as well as the quantum mechanical character of the electron capture process. For the quantum wells with no bound state close to the continuum, only a slight decrease of the carrier capture time as a function of laser energy is observed. We did not measure the capture times for laser excess energies larger than 41 meV, since for these laser energies, the electron excess energy exceeds 36 meV. As a result, the electrons are able to emit a LO phonon before the capture process, which complicates the distribution function.

Conclusions

We have measured the carrier capture time using two different experimental techniques: luminescence up-conversion measurements for measuring the rise-time of the quantum well luminescence and pump-probe correlation measurements for measuring the decay of the barrier population. Both techniques yield an oscillating carrier capture time which is also quantitatively in agreement with theory without any adjustable parameters.

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