RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 47, NUMBER 16 15 APRIL 1993-II

Ultrafast optical evidence for resonant electron capture in quantum wells

M. R. X. Barros and P. C. Becker
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

D. Morris, B. Deveaud, and A. Regreny
France Telecom, Centre National d'Etudes des Telecommunications, 22301 Lannion, France

F. Beisser
AT&T Bell Laboratories, Holmdel, New Jersey 07733
(Received 22 February 1993)

The dynamics of carrier capture in GaAs quantum wells have been measured with femtosecond resolution using pump and probe experiments under resonant excitation conditions. The electron capture mechanism at room temperature shows a resonance with a time as short as 650 fs for a well thickness of 58 Å. This gives strong evidence for the importance of the quantum-mechanical LO-phonon-assisted capture mechanism predicted by theoretical calculations.

The carrier capture time in quantum wells has been extensively studied in recent years due to both fundamental and practical interest in this process. The capture mechanism has been most recently considered within the framework of a quantum-mechanical description, where the fastest process is the scattering by LO phonons through the Fröhlich interaction. It has been predicted that this process should lead to strong resonances in the capture times of electrons, where the shortest capture times are on the 100-fs time scale.[1-3] Although luminescence experiments give evidence that such resonances may indeed exist,[4] to date, there have been no reports of direct measurements of a femtosecond oscillating carrier capture time in multiple quantum wells. Published experiments suggest that capture times are always short (less than a few ps).[5-7]

It has been proposed that for thick barriers or for high carrier densities, the coherence of the electron is not preserved long enough for a quantum-mechanical description to be valid, and therefore simple diffusion mechanisms apply.[8-10] In that case, the capture process only depends on the configuration of the barrier. A rather correct modeling of laser structure was indeed obtained with such a description. The possible validity of the quantum-mechanical description in the case of multiple-quantum-well (MQW) structure, when the period is less than the mean free path of the carriers, is a question of interest which has been raised previously without having received a clear answer. It is on this latter case that we focus our study.

In this paper, we present an investigation of capture mechanisms in $GaAs/Al_xGa_{1-x}As$ MQW structures. We have first studied a series of samples by luminescence with subpicosecond resolution and showed that the capture times were never longer than 3 ps, in contradiction with available theories. We performed quantum-mechanical calculations of the capture times and found that strong resonances are expected as a function of well width. We calculate times ranging between 80 fs and 20 ps for LO-phonon-assisted electron capture. The very short times have not been observed by luminescence techniques, due to the drawbacks of this technique as applied to carrier capture measurements in MQW's. We have performed, for the first time, to our knowledge, *femtosecond* pump and probe experiments to directly observe the carrier capture process. The carriers are resonantly excited at the bottom of the barrier, in contrast to previous luminescence experiments where the photoexcited carriers have a large excess energy. This eliminates relaxation processes which would otherwise obscure the capture process. Capture times in the few hundreds of femtosecond domain can now become observable.

The pump and probe experiments are done at room temperature. Thus, the calculated capture times, in first approximation, need to be averaged over the resulting thermal carrier distribution. This thermal average washes out the long capture times and the computed times are then found to be shorter than 3 ps for all well widths, in agreement with experiments. The model predicts a very short time resonance (120 fs) around 60 Å (for $x=0.3$ $Al_xGa_{1-x}As$). This corresponds to the resonant capture of an electron by emission of an LO phonon with very small wave vector between two states having a large wave-function overlap. Although we indeed observed in luminescence the shortest time at this position (approximately 1 ps), the very short times expected were not observed presumably because of competing relaxation channels.[11] A significantly shorter time was observed, however, when using the pump and probe technique.

We give here results of a pump and probe experiment where the excitation energy is adjusted to be resonant with the edge of the barrier states. Such a configuration prevents the effects of slow relaxation channels. The capture time of electrons is given by the recovery of the barrier absorption bleaching. Resonance in the electron capture time is clearly obtained in time-resolved experiments, and we demonstrate that this resonance indeed corresponds to a $q=0$ LO-phonon emission and a very short capture time of only 650 fs.

We have studied three samples of high quality grown

by molecular-beam epitaxy. The barrier thickness is 200 Å (for $x = 0.25$ Al) in all three samples in order to ensure the coherence of the electrons over at least one period of the MQW. The well widths are 39, 58, and 78 Å, with 20 barrier and well periods. The quality of the samples is checked by a combination of luminescence, luminescence excitation, and x-ray diffraction. The linewidth at low temperature is as narrow as 1.5 meV for the 78-Å sample. For the pump and probe measurements, the substrate is removed and the surfaces of the sample are antireflection coated with SiO. The room-temperature absorption spectra of the three samples are displayed in Fig. 1. For all three samples, the onset of the barrier absorption is observed at 720 nm. For the 39-Å sample, only one subband is evidenced. For the 78-Å sample, two of them are clearly resolved. The 58-Å sample is intermediate, as one shoulder can be observed just below the barrier, corresponding to a subband just bound into the well. This is exactly the condition for which resonant capture is expected.

The pump and probe measurements are made using a mode-locked Ti:sapphire laser operating at 720 nm with a pulse width of approximately 100 fs, amplified by a copper vapor laser pumped dye amplifier at a repetition rate of 8.5 kHz. The pump pulse has a spectral bandwidth of 25 meV. The pump intensity corresponds to an injected carrier density of approximately 10^{11} cm^{-2} in each well, and we have checked that the results do not depend on density in this range. In particular, renormalization or relaxation effects in the barrier can be neglected at such densities.

Typical pump and probe signals are shown in Fig. 2 for the 39-Å sample. The pump energy is resonant with the edge of the barrier, to avoid the observation of effect relaxation mechanisms in the barrier. The bleaching signal at the same energy decays with a short-time constant of 100–150 fs at early times followed by a longer time constant decay of about 2 ps as can be evidenced from the semilogarithmic plot of Fig. 3. In contrast, the bleaching signal at lower energies (in the well) rises with a time constant of about 2 ps as can be seen in Fig. 2(b). This corresponds to carriers progressively filling up the well as they are captured. Figure 3 compares the absorption bleaching curves at the barrier edge for the 39- and 58-Å well. The short-time decays are very similar; however, the long-time behavior is much more rapid in the 58-Å sample. Coherent artifact may only contribute in part to the short-time decay.

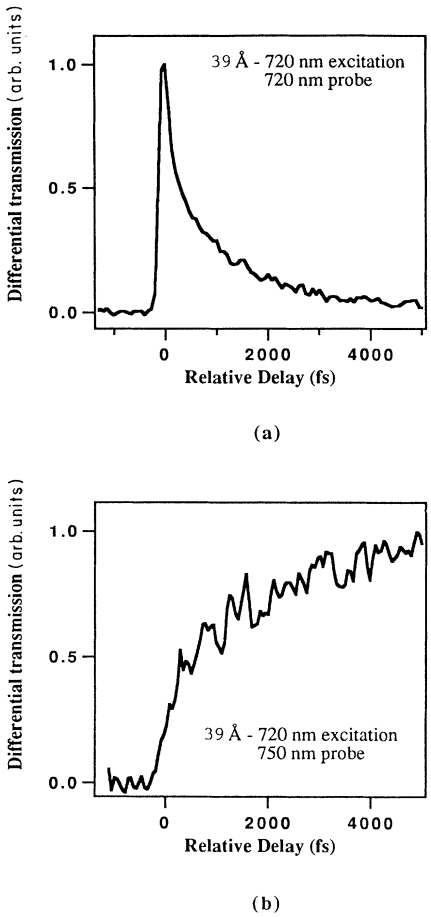

FIG. 2. Pump and probe differential transmission signal as a function of relative delay between pump and probe, for the 39-Å well, with pump and probe pulses centered at 720 nm (a) and with pump and probe centered at 720 and 750 nm, respectively (b).

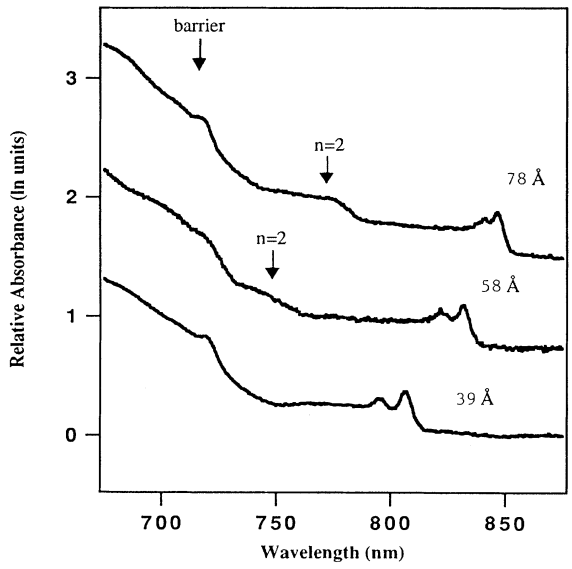

FIG. 1. Absorption spectra of the GaAs/$Al_xGa_{1-x}As$ MQW's used in this experiment, the well widths are 39, 58, and 78 Å. The curves for each well width have vertical offsets for convenience. The $n = 1$ and 2 arrows denote the onset of the first and second subband absorptions, respectively, and the barrier arrow denotes the onset of the $Al_xGa_{1-x}As$ barrier absorption.

The interpretation of our results is as follows. When exciting the sample at the energy of the barrier, we create at initial times electrons and holes in barrier states, and electrons and holes in well states (with large in-plane momentum). The short-time decay contains the relaxation of the carriers directly absorbed in the well. In fact,

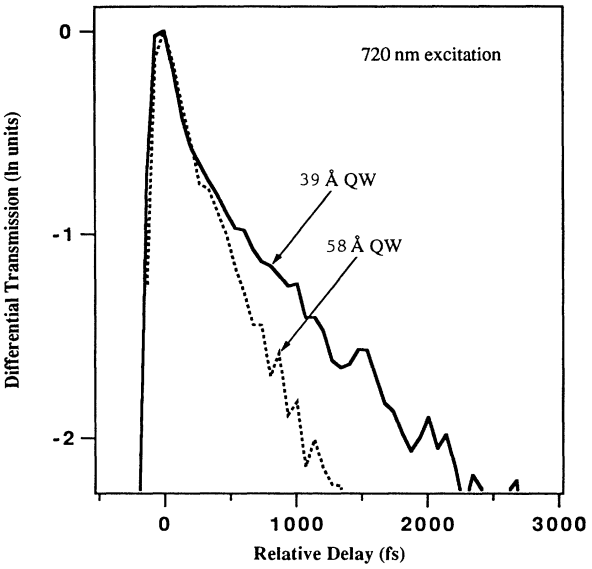

FIG. 3. Pump and probe differential transmission at 720 nm for the 39- and 58-Å wells, on a natural logarithm scale.

those carriers leave the optically active region within a characteristic time corresponding to the LO-phonon scattering time (i.e., 150 fs). Interestingly, the relative contribution of this part of the signal indeed corresponds roughly to the proportion of the carriers directly absorbed in the well. Figure 1 shows that in the 39-Å sample, the well absorption contributes about 30% of the total absorption at 720 nm whereas this contribution rises to about 70% in the 78-Å sample due to the presence of two subbands instead of one. The data for the 39-Å well indicate that about 40% of the signal relaxes with the first 150 fs whereas for the 78-Å well, the initial decay encompasses about 80% of the signal.

In principle, pump and probe experiments measure the sum of the contributions of the electrons and the holes, so that the slow decay in our experiment should reflect the contribution of both electrons and holes to the capture. One can argue, however, that in room-temperature experiments, the contribution of holes to the bleaching signal is negligible in first order. The excitation pulse has a bandwidth of 25 meV. It creates an initial distribution 20 meV wide for the electrons and only 5 meV wide for the holes, due to their different effective masses. After the carriers are injected at the band edge, they thermalize to the lattice temperature on a very fast time scale (150 fs) by electron-electron interaction and LO-phonon absorption. When thermalization occurs, both distributions spread over a Boltzmann distribution of width 25 meV, which does not affect greatly the optical contribution of electrons, but reduces dramatically the contribution of holes. Since the optically excited region is also the optically probed region for the degenerate pump and probe case, the holes, through thermalization, do not contribute to the long-time signal. The long delay time constant can then be attributed to the capture process of electrons. This statement is also supported by the fact that the resonance position is indeed the one expected for electrons, and not that for holes.

The decay times obtained in pump and probe (triangles) and in luminescence (squares) experiments are plotted in Fig. 4. A clear resonance is observed for a well width of 58 Å. There is a large relative difference between the pump and probe measurement and the luminescence measurement for the 58-Å sample, where the complex relaxation mechanisms in luminescence experiments preclude the observation of very short times. For the 39- and 78-Å wells the two measurements are in relatively good agreement.

On the same figure, we plot the results of a theoretical calculation of the electron capture times. The details of the calculation are published elsewhere.[11] This calculation involves the computation of the different levels and their wave functions in the envelope-function framework. Scattering with LO phonons via the Fröhlich interaction or with ionized impurities is included as a perturbation and bulk phonon modes are used in the calculation. The capture time calculated results from an average over the electron distribution. This model has no adjustable parameter. The impurity concentration, measured independently, was estimated to be about 2×10^{15} cm^{-2}. The calculation shows that the impurity contribution to the capture time is negligible at room temperature, at this concentration level (the impurity contribution is significant at temperatures less than 100 K).

The contribution of the temperature of the carriers is taken into account by a proper average over the initial states in the barrier. In order to perform the average, we assume that the distribution is thermalized at all times. In such a case, one can show that the decay of the whole population is the average of the capture time at each energy weighted by the Boltzmann factor. Comparison between the capture time computed for $T=0$ K (no thermal broadening of the electron distribution), and for $T=295$ K is shown in Fig. 5. Long capture times of about 20 ps are expected between 45 and 60 Å when the electrons cannot emit an LO phonon to be scattered to

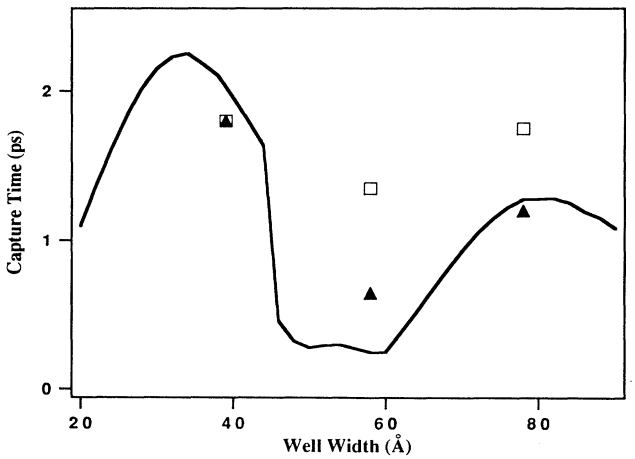

FIG. 4. Measured electron capture times (triangles, pump and probe; squares, luminescence) as a function of well width. The solid curve is the result of capture calculations including thermal averaging over the initial state distribution.

the $n=2$ level of the well. At 65 Å the $n=2$ level is just 36 meV below the barrier ground state and scattering becomes efficient again. At room temperature the high-energy part of the electron distribution can be scattered to this $n=2$ level very efficiently and the long times are washed out (solid curve) by the contribution of the hot electrons. At the same time, thermal averaging over the initial states makes the short-time resonances not as deep as in the 0-K model (250 instead of 80 fs). As can be seen from Fig. 4, the agreement between the theory for $T=295$ K and the experiment is very reasonable for a model with no adjustable parameters. The time experimentally observed for the 58-Å quantum well (650 fs) is longer than the calculated time (250 fs). This may be due to the overly simplistic approximation used in the thermal averaging. Indeed, thermal redistribution in the electron gas is not instantaneous; it may be assumed to occur with a characteristic time of the order of 100 fs, which is not very short compared to the capture time in the 60-Å well-width range.[12] Introduction of a finite thermalization time would certainly make the computed times longer.

In summary, we have observed that the electron capture times in MQW's are always very short (<2 ps) at room temperature. This arises from the fact that the electron energy distribution has a broad thermal width, and also from the fact that the thermalization time is fast. In this way, at room temperature, all the carriers will experience the effects of the fastest relaxation channel. We have observed that electron capture has a resonance around 60 Å, as predicted by the theory, with a capture time of 650 fs. This supports the picture of LO-phonon-assisted carrier capture, in which strong oscillations in the capture rate as a function of well width are expected.

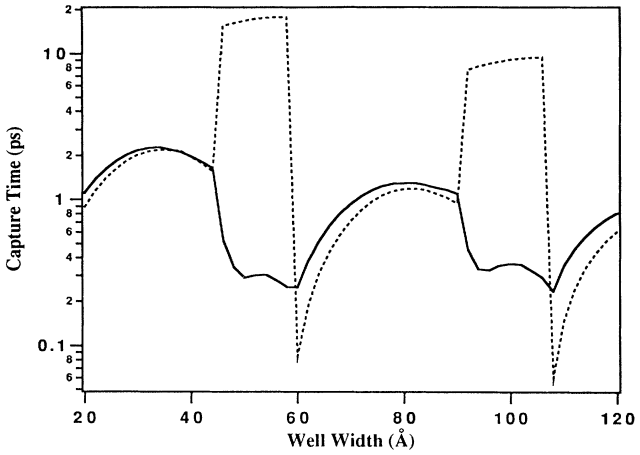

FIG. 5. Results of theoretical calculations. Dashed curve, LO-phonon-assisted capture for electrons at the bottom of the barrier; solid curve, average capture time for a thermal distribution of electrons at $T=295$ K starting from the bottom of the barrier.

We thank D. Chemla, A. Chomette, F. Clerot, G. Bastard, T. Damen, J. Shah, and J. Weiner for helpful discussions and P. Auvray for the x-ray characterization of the samples.

[1]S. V. Kozyrev and A. Ya. Shik, Fiz. Tekh. Poluprovodn. **19**, 1667 (1985) [Sov. Phys. Semicond. **19**, 1024 (1985)].

[2]J. A. Brum and G. Bastard, Phys. Rev. B **33**, 1420 (1985).

[3]M. Babiker and B. K. Ridley, Superlatt. Microstruct. **2**, 287 (1986).

[4]N. Ogasawara, A. Fujiwara, N. Ohgushi, S. Fukatsu, Y. Shiraki, Y. Katayama, and R. Ito, Phys. Rev. B **42**, 9562 (1990); P. W. M. Blom, C. Smit, J. E. M. Haverkort, and J. H. Wolter, *ibid.* **47**, 2072 (1993).

[5]B. Deveaud, J. Shah, T. C. Damen, and W. T. Tsang, Appl. Phys. Lett. **52**, 1886 (1988).

[6]R. Kersting, X. Q. Zhou, K. Wolter, D. Grutzmacher, and H. Kurz, Superlatt. Microstruct. **7**, 345 (1990).

[7]D. Y. Oberli, J. Shah, J. L. Jewel, T. C. Damen, and N. D. Chand, Appl. Phys. Lett. **54**, 1028 (1989).

[8]A. Weller, P. Thomas, J. Feldman, G. Peter, and E. Gobel, Appl. Phys. A **48**, 509 (1989).

[9]S. Morin, B. Deveaud, F. Clerot, K. Fujiwara, and K. Mitsunaga, IEEE J. Quantum Electron. **QE-27**, 1669 (1991).

[10]S. Weiss, J. M. Wiessenfeld, D. S. Chemla, G. Sucha, M. Wegener, G. Einsenstein, C. A. Burrus, A. G. Dentai, U. Koren, B. I. Miller, H. Temkin, R. A. Logan, and T. Tanbun-Ek, Appl. Phys. Lett. **60**, 9 (1992).

[11]B. Deveaud, D. Morris, A. Chomette, and A. Regreny, Solid State Commun. **85**, 367 (1993).

[12]T. Elssaesser, J. Shah, L. Rota, and P. Lugli, Phys. Rev. Lett. **66**, 1757 (1991).