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Density dependence of carrier-carrier-induced intersubband scattering in GaAs/$Al_xGa_{1-x}As$ quantum wells

M. Hartig, J. D. Ganière, P. E. Selbmann, and B. Deveaud
Physics Department, Institute of Micro and Optoelectronics, Swiss Federal Institute of Technology-EPFL, 1015 Lausanne, Switzerland

L. Rota
Ecce, 42019 Scandino, Italy

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Photoluminescence lifetimes of the $n=2$ level in a large quantum well show a clear nonmonotonic dependence on the density of optically generated carriers. Varying the power density over five orders of magnitude we prove directly the high efficiency of carrier-carrier interaction for intersubband scattering when longitudinal-optical phonon emission is suppressed. For low densities, the observed $n=2$ decay times get shorter (from 40 down to 5 ps) as the density is increased. At high densities Pauli blocking reduces significantly the intersubband scattering rates. [S0163-1829(99)06124-X]

Intersubband scattering (ISBS) in quantum wells (QW's) has been a subject of high interest for more than a decade, due to its importance as a basic process in fundamental semiconductor physics and also due to its high relevance in technical applications (intersubband lasers, infrared detectors).[1,2] ISBS in narrow QW's, where the electron subband separation between the first two confined subbands is larger than the energy of the longitudinal-optical (LO) phonon ($\Delta E_{1,2} > \hbar\omega_{\mathrm{LO}}$) is now well understood. ISBS is then governed by LO phonon emission and happens on a time scale of ≈ 1 ps.[3] Population inversion, although possible to achieve,[1] needs careful band gap engineering. Large QW's on the contrary, where $\Delta E_{1,2} < \hbar\omega_{\mathrm{LO}}$, seem to promise a high potential for population inversion. Indeed, much longer lifetimes of the $n=2$ electron population were suggested because, in such conditions, LO phonon emission is suppressed.[4] The results on ISBS in wide QW's remain however very contradictory; published scattering times vary between 1 ps and 1 ns. [5–12]

In samples with subband separations below the LO phonon threshold, ISBS can only be due to acoustic phonon emission and carrier-carrier (CC) interaction. While longitudinal-acoustical phonon scattering is inefficient for ISBS (due to the long scattering time of more than 100 ps and the small energy removal of ≈ 1 meV),[4] CC scattering has been underestimated for a long time as a relevant ISBS process, despite the fact that some theoretical calculations showed more than ten years ago that it should be quite efficient.[13] Recently the importance and efficiency of CC scattering for ISBS in large QW's could be directly evidenced, in good agreement with Monte Carlo simulations.[12] At a density of about 10^{11} cm^{-2} the decay of the $n=2$ electron population in a large QW was found to be of the order of a few ps only.

It is well known, that CC interaction plays an important role for the relaxation dynamics in semiconductors. Energy exchange within a given subband is a very fast process and happens in most cases on a subpicosecond time scale.[14] It redistributes the carriers according to a Fermi distribution. As a result one observes $n=1$ photoluminescence (PL) at the shortest times and the spectra display an exponential slope on the high-energy side, indicating a thermalized carrier distribution. Interaction between carriers confined in different subbands also leads to exchange of an energy and momentum, driving the carrier populations towards a quasiequilibrium. The most prominent CC processes for interaction between the first and the second electron subband are labeled 1212, 2112, and 2211 (process $klmn$ describes an interaction, where charge carrier 1, initially in state k scatters to m under collision with partner 2, which scatters from l to n).

Processes 1212 and 2112 cause exchange of energy and momentum of electrons located in different subbands, leading to temperature equality between electron populations located in different subbands. The total number of electrons in each subband remains unchanged. In process 1212 electron 1, initially in $n=1$ is transferred to another state in $n=1$, while electron 2 scatters between two states in $n=2$, under conservation of energy and momentum. By 2112 the electrons from $n=1$ and $n=2$ scatter to the other subband without changing the number of electrons in each subband (see Fig. 1). The characteristic time for the establishment of a common temperature depends on carrier density, and typical time scales are between 1 and 10 ps for densities between 10^{10} cm^{-2} and 10^{11} cm^{-2}. [15]

Process 2211, on the other hand, is the most effective ISBS process when LO phonon scattering is forbidden and for densities around 10^{11} cm^{-2}. Under conservation of energy and momentum two charge carriers, initially in $n=2$ scatter to $n=1$, whereby one of the particles looses a certain amount of energy, which is gained by the second carrier. The scattering times can even be subpicosecond at these densities.[12] Energy redistribution through 1111, and cooling then follows within the $n=1$ subband.

1212 scattering has an efficiency roughly one order of magnitude larger than 1221 and 2211.[13] It is, however, important to notice that ISBS by process 2211, whereby two electrons leave the $n=2$ subband after a single collision, is possible without any transfer of energy (see Fig. 1). Establishment of temperature equality between the two subbands,

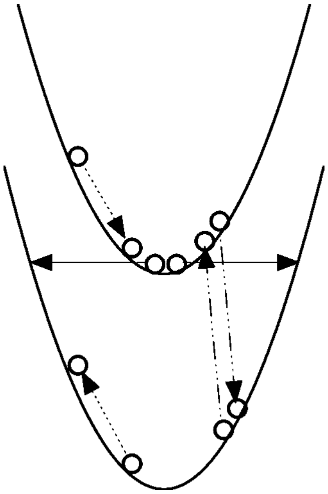

FIG. 1. Dominant intersubband CC interaction processes in a quantum well. Intersubband transition [2211] (full line), intersubband thermalization [1212] (dotted line), and [2112] (dash-dotted line).

on the contrary, requires energy transfer form the higher to the lower subband. This happens by much more than one collision per electron, as the energy transferred per collision is quite small. Furthermore, this exchanged energy decreases as the carriers accumulate closer to $k=0$ (small number of carriers, low temperature). Thus, although F_{2211} is 10 times smaller than F_{1212}, we can expect that ISBS due to CC interaction and temperature equalization between subbands will happen on comparable time scales. With decreasing subband separation the CC-ISBS rate increases due to the reduction of the exchanged wave vector (a bulk system can be considered as the limit of a very wide QW, where CC scattering times are very short[12]).

In this paper we give a direct proof that ISBS is indeed due to CC scattering. Tuning the excitation density between 5×10^{7} cm^{-2} and 2×10^{11} cm^{-2} the $n=2$ lifetimes vary from 40 to 5 ps. This confirms the very large efficiency of CC scattering for ISBS. When the density is further increased, the $n=2$ lifetime increases again. This is due to band filling in $n=1$ and Pauli blocking.

We use photoluminescence as this allows us to determine directly, and with high sensitivity, the dynamics of the generated charge carriers in time- and energy-resolved measurements. A Ti:sapphire laser is used for the excitation of the samples and PL is detected with a synchroscan streak camera. In order to narrow the width of the spectrally broad 100 fs pulses, a 3 nm bandpass filter has been used. According to the bandwidth of the filter (5.9 meV) the pulse width was 400 fs. The overall time resolution of the experiment is 5 ps, if an optical grating of 150 lines per mm (l/mm) is used in the monochromator. For a better spectral resolution of 2 meV some of the measurements have been performed using a grating of 600 l/mm, however with the drawback of lower time resolution. The sample is immersed in superfluid helium for the measurements at 2 K. The measurements at high power density ($>2\times10^{11}$ cm^{-2}) were performed by imaging the luminescence spot and spatially filtering it with a 50 μm pinhole, in order to select only the central part of the excited spot, over which the excitation density is homogeneous. We investigated a series of high-quality asymmetric coupled well Al-Ga-As/GaAs samples. They consist of a wide QW (WW) of 210 Å, coupled to a narrow QW (NW) of 90 Å through a barrier of 40 Å. The coupled well pairs are separated by 100 Å barriers of Al-Ga-As, with 25% of Al.

Due to the high quality of our molecular-beam epitaxy samples, the lifetime of the carriers in the $n=1$ subband is longer than the time between two successive laser pulses (12 ns). As a consequence, a sizeable number of cold carriers is still present in the QW, when a new population of hot carriers is excited by the next laser pulse. The number of remaining cold carriers is often found to be more than 10% of the total number of generated carriers. Due to the fast interaction and energy exchange between the cold ''old'' and the hotter ''new'' carrier population by intraband and intersubband interactions, ISBS and thermalization dynamics are accelerated in an unpredictable way. In order to empty the $n=1$ subband faster the samples have been slightly irradiated by high-energy electrons (2 MeV, 10^{14} cm^{-2}). This creates recombination centers in the bandgap, and reduces the lifetime through nonradiative recombinations. Compared to ISBS times the lifetime remains nevertheless long (>1 ns), due to the moderate number of recombination centers.

The PL spectra give direct information on the distribution function of the charge carriers. As already described in detail in Ref. 15 the investigation of ISBS has to start from a nonthermalized carrier distribution (different carrier temperatures in the subbands T_i and different chemical potentials μ_i). Otherwise the observed decay of the $n=2$ population is only due to cooling of the overall population in the QW and not due to ISBS.[13]

As has been shown recently, ISBS scattering due to CC interaction is very fast and, therefore, very difficult to observe in regular QW samples. To the best of our knowledge no spectrum showing nonthermalized $n=1$ and $n=2$ subbands has ever been reported for a WW. Such nonthermalized distribution can be more easily observed in an asymmetric coupled QW (CQW). The overlap of the corresponding wave functions through the thin barrier leads to coupled states that are in first approximation linear combinations of the uncoupled QW levels: $\Psi=(1/\sqrt{2})*[\Psi_A(n=2)\pm\Psi_B(n=1)]$. Evaluation of the matrix element for the probability of an intersubband transition between the $n=2$ and $n=1$ subband in the wide QW results in a lifetime for the $n=2$ population exactly twice as long as in a single QW. This lower limit for the ISBS time is obtained in the case of a perfect resonance.[17] As the density of states contributing to the $n=2$ subband is now larger compared to a single QW the PL intensity will be stronger too. In the specific sample used in this study the coupling of the energy levels, which have identical confinement energies in the uncoupled wells, results in a splitting of 5 meV between the two transitions.

Typical sets of PL spectra at different delay times are shown in Fig. 2. The spectra have been obtained by summing over 20 pixels along the time axis. In order to create a cold exciton population in the coupled levels, the laser has been set in resonance with the lower of the split levels. At early times the strong peak at 1.553 eV is dominated by backscattered light of the laser, while after ≈20 ps a pure PL signal of the $(e,hh)_2$ transition in the WW is observed. The PL maxima at energies of 1.523 and 1.525 eV are due to the $(e,hh)_1$ and $(e,lh)_1$ exciton transitions in the WW. Using a

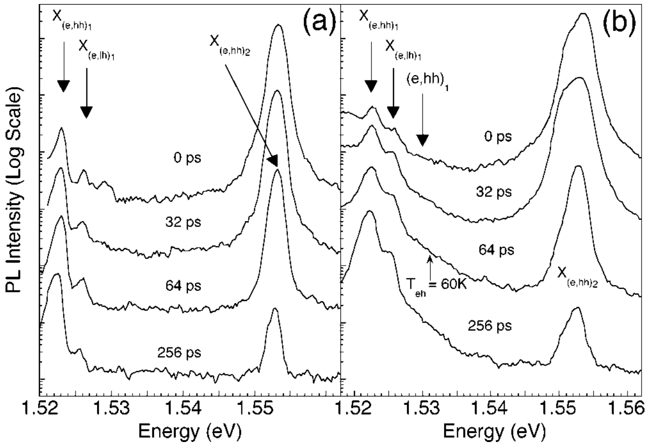

FIG. 2. Luminescence of a 90/40/210 CQW at different delay times. The excitation densities are (a) 4.2×10^{8} cm^{-2} and (b) 1.3×10^{10}cm^{-2}. The transitions are mainly excitonic in the low density regime, and free carriers may be observed in the spectra recorded at higher densities.

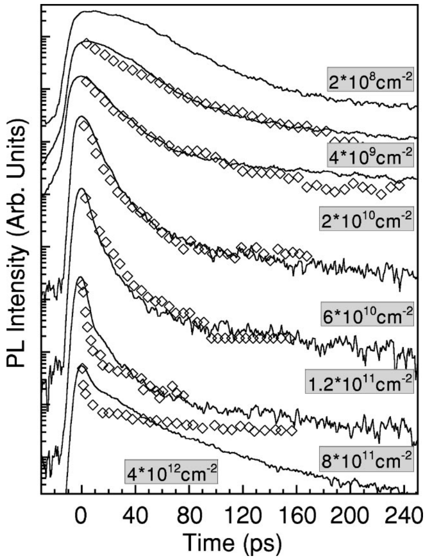

FIG. 3. Time-resolved transients of PL at the energy of the $n=2$ transition in the WW of a 90/40/210 CQW sample. The decay times shorten with increasing carrier density due to stronger CC interaction and are below the time resolution in the transient at 1.8×10^{11} cm^{-2}. At higher densities increasing decay times are due to band filling. The diamonds represent the results of a Monte Carlo simulation with no adjustable parameter.

higher excitation density [Fig. 2(b)] causes visible occupation of the $n=1$ continuum states. The carrier population in the lowest subband thermalizes within the time resolution of the experiment and an exponential slope on the high-energy side of the $(e,hh)_1$ transition can be observed at the earliest times. Excitonic and free carrier populations equilibrate at long times according to the mass action law.[18] However, the spectra demonstrate clearly the dynamics of the charge carriers, which are directly generated high in the $n=1$ subband or get there after ISBS, and relax to the bottom of the QW by intrasubband collisions and cooling.

The signal corresponding to the $(e,hh)_2$ transition is maximum at $t=0$ and then decreases within 100 ps. During the same period, the PL intensity of the $n=1$ transitions (excitons and free carriers) of the WW builds up before decaying for longer times. The first part of this evolution represents ISBS and cannot be explained by a cooling process. In the case of thermal equilibrium between $n=2$ and $n=1$ populations, the relative intensity of the PL maxima would be observed according to a Maxwell-Boltzmann statistics. PL maxima in $n=2$ and $n=1$ are then related by a factor $\exp(\Delta E/k_BT)\times$DOS (DOS is the joint density of states). Such a situation is never established in any of our spectra at delay times shorter than 100 ps. LO phonon emission by electrons in $n=2$ can safely be excluded as a possible mechanism for ISBS, as the excitation energy is well below the corresponding threshold at 41 meV above the $(e,hh)_1$ excitonic transition.

We emphasize that only electron dynamics is observed in the present experiments, even though the luminescence intensity depends on the presence of both kinds of carriers. As has been shown in (Ref. 16), hole intrasubband thermalization happens on a much shorter time scale than electron thermalization due to more efficient CC scattering (because of the larger effective hole mass) and due to the smaller initial excess energy of the holes. As the separation of the first two hole subbands is much smaller than for electrons, interband thermalization as well as ISBS can safely be assumed to happen even faster compared to electrons. This is confirmed by our Monte Carlo simulations. This justifies that, in our simple description, we do not consider hole redistribution as it occurs beyond our time resolution.

Figure 3 displays the decay curves at 1.553 eV for different excitation densities. In order to obtain the best temporal resolution, a 150 l/mm grating has been used. The curves evidence the strong density dependence of CC interaction induced ISBS: At low carrier densities the CC scattering is not very efficient and the $n=2$ decay times are of the order of 35 ps. Below approximately 10^{9} cm^{-2} the $n=2$ decay time saturates at about 35 ps (Fig. 3). Increasing the excitation density intensifies the CC interaction and causes shorter decay times. Above 5×10^{10} cm^{-2} the ISBS times get shorter than the temporal resolution of the experiment of 5 ps (transient at 1.8×10^{11} cm^{-2}). At very high densities, longer time decays can be identified in the transients, but the luminescence is hidden, for the duration of the laser pulse, by light scattered from the sample surface. Comparison of the decay curves with Monte Carlo calculations show a very good agreement, and allow to confirm that holes indeed have a negligible effect on the observed dynamics.

Figure 4 shows the ISBS times over five orders of magnitude. Three regimes can be defined: At the lowest densities ($>1\times 10^{9}$ cm^{-2}) ISBS is independent on power density due to inefficient CC interaction and determined by other processes. Optical recombination of $(e,hh)_2$ excitons has to be taken into account for the removal of carriers from $n=2$ in the WW. It is difficult to make a definite statement whether the dynamics of free carriers or correlated electron hole pairs is observed in the very low density regime. It is possible that the very short times observed here are due to the enhanced radiative recombination of cold $n=2$ excitons, similar to what has been observed for $n=1$ excitons.[19] This, however, does not change any of our considerations on ISBS. For power densities between 1×10^{9} cm^{-2} and 2

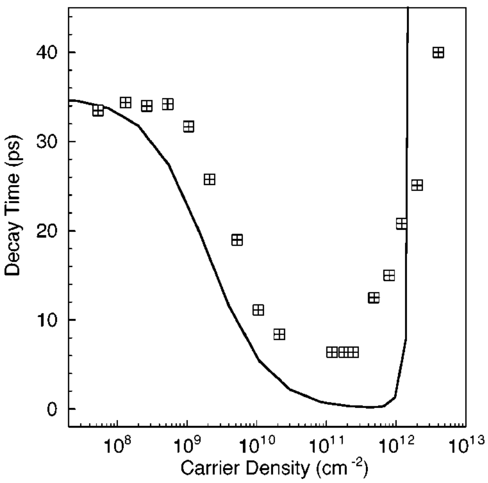

FIG. 4. Experimental $n=2$ PL decay times of a WW the $n=2$ luminescence in a wide QW (crossed squares). A calculation confirms the experimental findings (solid line) of ISBS due to CC interaction. The flatening at low densities is due to an additional recombination time of 34 ps, presumably due to radiative coupling.

$\times 10^{11}$ cm^{-2} ISBS is clearly governed by CC interaction. Decay times shorten by about a factor of 10 with increasing power density, according to the enhanced scattering probability. In the high density regime above 2×10^{11} cm^{-2} phase space filling prevents ISBS due to Pauli blocking.

The experimental findings are compared with a calculation evaluating the intersubband matrix element for the CC scattering process 2211. A simultaneous process removing electrons from $n=2$ with an exponential time constant of 35 ps has been assumed at low densities, leading to good correspondence with the experimental data. This time constant is possibly due to radiative recombination of excitons. Pauli blocking, introduced very directly by the Fermi filling of the final states, is well evidenced in the calculation and responsible for the steep rise of the curve for densities $>10^{12}$ cm^{-2} (solid line).

In conclusion, we have observed directly the dependence of ISBS times on density of carriers in a wide QW, in a density range from 5×10^7 cm^{-2} to 5×10^{12} cm^{-2}. Due to the small subband separation, ISBS due to LO phonon emission was forbidden. With increasing densities, ISBS times were observed to shorten significantly. This undoubtedly confirms a CC-induced ISBS process: *Only stronger CC interaction can explain shorter scattering times with increasing density*. The contrary is expected for a cooling process. Longer decay times of $n=2$ electrons at high densities are due to Pauli blocking in $n=1$. The experimental results are in very good agreement with both a simple calculation of carrier-carrier scattering and a full Monte Carlo simulation.

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