Efficient Intersubband Scattering via Carrier-Carrier Interaction in Quantum Wells

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(Received 12 August 1997)

Using femtosecond resonant luminescence, we have measured the intersubband scattering rate of electrons in wide GaAs quantum wells at very low excitation densities. Even when the spacing between the first two electron subbands is smaller than the LO phonon energy, we observe that intersubband scattering is a subpicosecond process, much faster than previously measured or anticipated. Our experimental results are in perfect agreement with Monte Carlo calculations, which show that carrier-carrier interaction is responsible for the ultrafast transitions. [S0031-9007(98)05430-1]

PACS numbers: 73.20.Dx, 72.20.Dp, 78.47.+p, 78.55.Cr

The dynamical properties of charge carriers in semiconductors are governed by Coulomb interactions and crystal lattice excitations. Energy relaxation of carriers happens on ultrashort time scales. Reduced dimensionality in artificial semiconductor systems can strongly influence the dynamical relaxation properties, as energy transfer is often possible only in quantized fashion. After extensive investigations during the past decade it is generally accepted that longitudinal optical (LO) phonon emission is the most efficient intersubband scattering (ISBS) process (time scale of $\approx$1 ps [1–3]). Depending on the size of the quantized system, the energy separation between the first two confined subbands can be above or below the LO phonon energy ($\hbar\omega_{\mathrm{LO}}$), allowing or suppressing LO phonon emission.

Wide quantum wells (QW), in which LO phonon emission is suppressed, offer a greater potential for population inversion, which is important for the design of light emitting devices (e.g., quantum cascade laser [4]). ISBS in those QWs has received much attention, leading to contradictory results [5–11]. This is due mainly to the difficulty of carrying out accurate experiments in wide QWs. Ultrafast experiments require lasers with a broad energy spectrum ($\approx$20 meV) resulting in broad carrier populations. When the energy separation between the first two subbands is smaller than $\hbar\omega_{\mathrm{LO}}$, many carriers may be excited above the threshold energy and can thus relax to the first subband through LO phonon emission. This effect can be reduced by lowering the initial laser energy, but this results in a relatively small fraction of carriers being excited in $n = 2$. At densities sufficiently high to obtain a signal from $n = 2$, Pauli blocking starts to play a major role reducing strongly all ISBS rates. In addition, it is not sufficient for a reliable study of ISBS to monitor the electron population at one given energy, but instead the dynamics of the entire distribution functions has to be probed. This explains why most of the results obtained up to date by different groups are rather conflicting. Using time-resolved techniques [5–7] scattering times between 20 and 750 ps were measured. Indirect steady-state techniques [8–11] also gave contradictory values, varying from 1 ps to 1 ns.

In this Letter we present measurements of ISBS in wide QWs ($\Delta E_{1,2} < \hbar\omega_{\mathrm{LO}}$), using resonant photoluminescence (PL) upconversion with 150 fs time resolution and under conditions where LO phonon emission is almost totally suppressed. Optimization of the experimental setup and a special sample design allow us to work with densities of optically generated carriers below 1×10^{10} cm^{-2}, i.e., more than a factor of 10 smaller than in any other experiment. We will show that carrier-carrier (CC) scattering is a much more efficient energy relaxation channel than previously expected and leads, under usual conditions, to ISBS times always shorter than 10 ps. This controversy and the relevance for applications, plus the fact that the importance of CC scattering for ISBS has been neclected so far, make our conclusions of wide interest. The discrepancy among published results will then be explained by Pauli blocking effects.

Unlike other techniques, when using proper experimental conditions, PL upconversion allows one to get directly the dynamics and the distribution function of charge carriers down to very low excitation densities. Suppression of LO phonon emission requires the injection of a cold enough carrier population in the $n = 2$ subband [12]. An experimental setup with two synchronously pulsed laser beams of different wavelengths makes it possible to generate charge carrier pairs resonant with the $n = 2$ electron-heavy-hole $(e, hh)_2$ transition and detect PL at the same frequency [13].

We have shown in former experiments in narrow QWs ($\Delta E_{1,2} > \hbar\omega_{\mathrm{LO}}$) [2] that excitation density plays a crucial role in the dynamics of ISBS. Energy exchange among electrons within a given subband is always a very fast process (faster than the time resolution of PL experiments (<100 fs) [14]). As a result, $n = 1$ PL is observed at the shortest times and PL spectra display an exponential slope on the high energy side, indicating a thermalized carrier distribution. Energy and carrier exchange between different subbands, however, depend decisively on excitation density. If only a small number of carriers is excited, different electron temperatures are observed in the two lowest subbands up to long delay times (≈4 ps) [2]. At higher densities ($\approx 3 \times 10^{11}$ cm^{-2}) we observe clearly that, at times as short as 500 fs, the carrier temperature is the same in the two subbands and the population ratio equilibrates according to this temperature. The decay of the $n = 2$ PL then corresponds only to cooling of the whole population and not to ISBS. These observations already indicate clearly the importance of CC induced energy averaging between two subbands with characteristic times that can be very short.

In addition to equal temperatures, thermal equilibrium requires a common chemical potential, μ_i, for both subbands. We find that this carrier exchange occurs on the same time scale as the equalization of the temperatures among the subbands. The investigation of ISBS has therefore to start from a nonthermalized carrier distribution (different T_i and μ_i) which has to be monitored as a function of time. In wide QWs no report exists, to our knowledge, of a time-resolved spectrum with clear indication of a nonthermal carrier distribution in the $n = 2$ subband, characterized by different carrier temperatures and/or chemical potentials in the $n = 1$ and $n = 2$ subbands. PL spectra allow for a direct display and determination of the electron and hole distribution functions. For example, after achieving thermal equilibrium, the PL intensity jumps by a factor of 2 at the onset of the $(e, hh)_2$ transition, according to the joint density of states. Limitations are given by spectral broadening due to the 150 fs gating pulse (11–14 meV here) and by the relatively low signal of the $(e, hh)_2$ transition, which requires a careful optimization of the detection sensitivity, as achieved here.

Now we address the question of how the $n = 2$ population decays to equilibrium in wide QWs. We show in Fig. 1(a) the spectrum of a GaAs/Al$_x$Ga$_{1-x}$As sample with a 210 Å QW obtained with a density of 3.5×10^{10} cm^{-2} carriers, after a 600 fs delay. The separation between the $n = 2$ and $n = 1$ electron subband is only 28 meV and carriers are generated resonantly with the $(e, hh)_2$ transition at 1.548 meV, to minimize LO phonon emission. The exponential decay between 1.52 and 1.54 eV indicates a thermalized population in the $n = 1$ electron subband. The $(e, hh)_2$ transition appears in the spectrum, even at this early time delay of 600 fs, only as a minor modification of the high energy tail. In fact, the PL spectrum is due to a fully thermalized carrier distribution in both subbands (equal μ_i's), the expected step of two at the onset of the $(e, hh)_2$ transition being broadened due to the femtosecond resolution. The open circles simulate such a thermalized spectrum convoluted by the gating pulse spectrum. The perfect agreement with the experiment proves that both subband populations have already been brought to thermal equilibrium. The thermalized electron population in $n = 2$ leads us to the conclusion that a very rapid scattering and energy exchange between the two subbands must have already happened within 600 fs. The same result has been obtained on a number of samples with well widths above 200 Å.

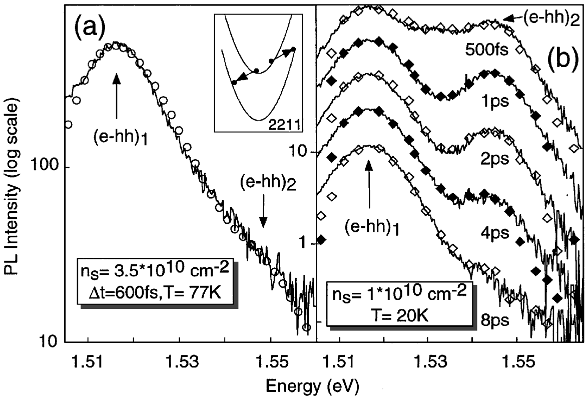

FIG. 1. (a) Spectrum of a 210 Å GaAs MQW sample at $\Delta t = 600$ fs. Only a small hint of the $n = 2$ subband can be observed at about 1.55 eV. Circles: calculation of the PL spectrum under conditions of thermal equilibrium between electrons in $n = 2$ and $n = 1$. (b) The PL spectra of a CQW sample at various times after the laser pulse. The diamonds represent the results of the Monte Carlo simulation. Inset: Most important CC-ISBS process (2211).

We then designed samples where the contribution of the $n = 2$ subband would be easier to observe. This has been done with a set of coupled asymmetric QWs (CQW): A wide QW coupled to a narrow one through a narrow barrier. Samples containing CQWs of GaAs with widths of 220 Å (well A) and 90 Å (well B) have been grown. The confinement energies of the electron levels $n = 2$ in the wide QW and $n = 1$ in the narrow QW are identical. The overlap of the corresponding wave functions through the 30 Å barrier leads to two eigenstates that are, in first approximation, linear combinations of the uncoupled QW levels: $\Psi = (1/\sqrt{2})[\Psi_A(n = 2) \pm \Psi_B(n = 1)]$. Evaluating the matrix element for the probability of an intersubband transition between the $n = 2$ and $n = 1$ subbands in the wide QW results in a lifetime for the $n = 2$ electron population exactly twice as long as in a single wide QW [15]. 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 also. Moreover, in order to be able to perform the measurements with the lowest possible carrier density, the sample was p doped [16].

We show in Fig. 1(b) a log plot of the PL spectra of one of the CQW samples at different delay times after the laser pulse. The maximum of the exciting laser (22 meV spectral width) was set at 1.532 eV, slightly below the $(e, hh)_2$ transition, to generate a carrier population as cold as possible (in the trace taken at 500 fs a hint of scattered light of the laser pulse can still be observed). The excited carrier density was 1×10^{10} cm^{-2} and the lattice temperature was 20 K. These spectra clearly show two peaks at 1.515 and 1.545 eV that correspond to the $(e, hh)_1$ and $(e, hh)_2$ transitions, respectively [17]. Over a time scale of a few ps, the PL intensity of the $(e, hh)_2$ transition vanishes and, simultaneously, the $n = 1$ signal increases [18]. To our knowledge this is the best direct evidence of ISBS in a sample with $\Delta E_{1,2} < \hbar\omega_{\mathrm{LO}}$. The ultrafast decay of the $n = 2$ peak indicates that the ISBS process is indeed very efficient even under these conditions.

In Fig. 2, the temporal evolution of the PL intensity (solid line) at the $(e, hh)_2$ transition energy of 1.545 eV for the CQW sample is presented for different excitation densities. Sum-frequency generation with the scatter of the laser pulse results in a strong peak at $t = 0$, demonstrating the time resolution of the experiment. At later times the signal is due to PL and shows an exponential decay for all densities with a time constant depending on density. At a density of (5×10^{10} cm^{-2}) the decay time of the $n = 2$ population is 2.9 ps; it remains constant for lower densities. If we assume perfect matching of the coupled levels, it follows that the ISBS time in a simple QW must be half this value. This is an upper limit if the levels are not matched. We must conclude that the $n = 2$ population in a single 220 Å QW decays to equilibrium with the $n = 1$ population within a time shorter than 1.5 ps. This is the shortest ISBS time measured directly for a sample where $\Delta E_{1,2} < \hbar\omega_{\mathrm{LO}}$. With increasing carrier density we can observe a systematic increase in the ISBS time.

To analyze the experimental results we made use of a Monte Carlo (MC) simulation which allows one to model easily the particular sample structure and to insert the relevant scattering mechanisms. We considered a parabolic band model, because the region of interest is quite close to the band gap and included LO and longitudinal acoustic (LA) phonons. Finally, CC interaction (electrons and holes) was included within the Born approximation and a static screening approach [19], which has been shown to give a good description of the phenomena in similar conditions [1,2,14]. Both the time and the energy resolved spectra were obtained, upconvolving the distribution function with the laser pulse, exactly as it happens in the experiments. The open diamonds in Fig. 1(b) represent the spectra obtained from the MC simulation at the corresponding times. From the excellent agreement we can deduce that the theoretical model represents the physical scenario very well.

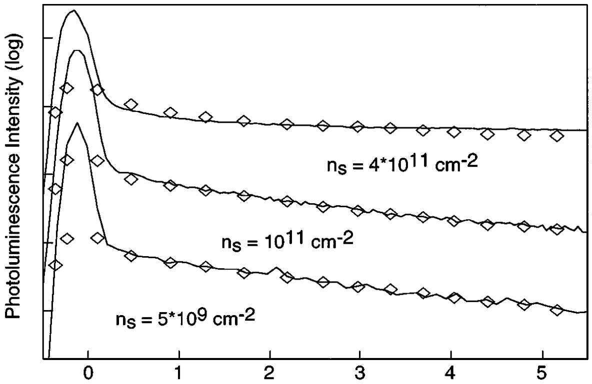

FIG. 2. Time evolution of the luminescence signal at 1.545 eV in the CQW of Fig. 1(b), corresponding to the $(e, hh)_2$ transition for three different excitation densities (solid line). The experimental results are well reproduced by the MC simulation (open diamonds).

A careful evaluation of both the experimental data and the MC results allows us to extract significant additional information. The mean energy of the carriers in the $n = 2$ electron subband does not decrease with time, but actually increases slowly during the first picoseconds. As a result, the exponential slope of the high energy tail of the second subband becomes less steep.

This heating gives us some insight into the CC scattering mechanism. In the inset of Fig. 1 is shown the most effective ISBS process due to CC interaction. Two electrons g and h, initially in $n = 2$, scatter to states i and j in $n = 1$ ($ghij = 2211$). While the first electron loses a certain energy, the collision partner gains exactly the same amount. As the CC scattering rate increases for small exchanged wave vectors, it is clear that preferably electrons with $K = 0$, i.e., those with very little kinetic energy, are scattered to $n = 1$. A number of further but, however, less efficient CC interaction processes, e.g., (2231), are possible leading to a net transfer of electrons. The MC simulation shows, furthermore, that approximately 25% of the carriers in $n = 2$ are still scattered to $n = 1$ through the emission of LO phonons. These are electrons in the $n = 2$ subband that have gained enough energy after a CC collision to emit an LO phonon. However, this process is not sufficient to cool the population in $n = 2$ as proposed by other authors [6,12,20], because the larger fraction of the carriers in $n = 2$ (75%) scatters to $n = 1$ through CC scattering with a significant increase of the second subband carrier temperature.

In (2112) an electron scatters from $n = 2$ to $n = 1$, while the partner particle scatters from $n = 1$ to $n = 2$ with the number of electrons in $n = 2$ remaining unchanged. According to its form factor this process is as fast as (2211) [21] and causes an exchange of energy between the subbands leading to temperature equalization. Electron-hole interaction processes of comparable efficiency are also possible and induce ISBS of electrons from $n = 2$ to $n = 1$. Electron-hole interaction and the p doping fixes the ISBS times to 2.9 ps for excitation densities below 5×10^{10} cm^{-2}.

Our calculations also explain a number of interesting results. ISBS due to CC interaction is more and more efficient as the well width increases (reduction of the

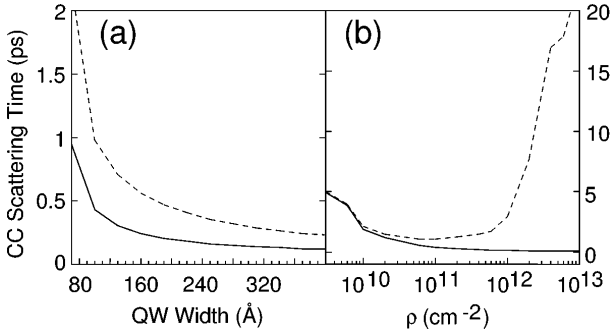

FIG. 3. CC-ISBS time for transitions $n = 2$ to $n = 1$, with (dashed line) and without (solid line) the Pauli blocking effect (a) as a function of the well width, at a fixed density of 5×10^{11} cm^{-2}, and (b) as a function of the density, for a 210 Å QW sample, also including the CC-ISBS from $n = 1$ to $n = 2$.

exchanged wave vector). This can be understood if we consider a bulk system as the limit of a very wide QW, where typical CC scattering times are of the order of 100 fs [19]. Moreover, this is in perfect agreement with our observations of the thermalization behavior as a function of QW thickness. When the electron subband spacing is of the order of 100 meV or more, LO phonon scattering is 10 times more efficient than CC scattering for densities of a few 10^{10} cm^{-2}. For wider QWs the contribution due to CC becomes almost equal to LO phonon scattering. Figure 3(a) shows the CC-ISBS time from $n = 2$ to $n = 1$ as a function of the QW width at a density of 5×10^{11} cm^{-2}. The solid line corresponds to the CC-ISBS time without considering the Pauli exclusion principle and indicates a decrease in time as the QW gets wider. When Pauli blocking is included the scattering time is reduced because of subband filling in $n = 1$.

Our results agree with some other experiments: Levenson *et al.* [6] measure a ISBS time of 20 ps, limited by their temporal resolution in a QW of 225 Å, Helm *et al.* [8] obtain a time of 1–2 ps from a theoretical fit on their saturation experiment. If the QW width is changed, for example, from 80 to 400 Å, the density at which filling strongly influences the decay times of the $n = 2$ electron population reduces by at least 1 order of magnitude. This makes experiments with large QWs and avoiding Pauli blocking difficult [Fig. 3(b)]. In other experiments either the time resolution was not good enough to observe the very fast early transient or the excited density was too high.

Finally, we can then consider which consequences these results have for possible intersubband laser applications. In order to obtain laser action, a reasonable population of the $n = 2$ state has to be obtained. According to our results this leads to fast CC-ISBS for densities where Pauli blocking is not yet effective. Contrary to the common belief, population inversion is therefore not easier to achieve in wide QWs than in narrow ones.

In conclusion, we observed fully thermalized luminescence spectra (electrons and holes) after 1 ps in SWQs and after 8 ps in CQWs. This gives an ISBS rate larger than 5×10^{11} s^{-1} due to CC scattering in wide QWs ($\Delta E_{1,2} < \hbar\omega_{\mathrm{LO}}$), evidencing the high efficiency of e-e and e-h interaction. A careful analysis of the results using Monte Carlo simulations allows us to understand the ISBS process and its density dependence in great detail. Most of the ISBS under these conditions is caused by CC interaction. ISBS may be dominated by LA phonons only in the limit of very low densities ($<10^9$ cm^{-2}) and in the absence of p doping. Finally, the strong dependence of the ISBS on excitation density in a relatively narrow range explains the experimental difficulties and the large scatter of values obtained in previous experiments.

We wish to thank A. Regreny and F. Morier-Genoud for growing the samples used in this study. We benefited from numerous discussions with D. Oberli, I. Bar-Joseph, and D. Morris. This work is financed by the Swiss FNRS under Contract No. 21-40345.94 and by OFES under 930332.

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