Femtosecond luminescence measurements of the intersubband scattering rate in $Al_xGa_{1-x}As/GaAs$ quantum wells under selective excitation

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We have investigated the intersubband scattering of electrons in GaAs quantum wells using luminescence up-conversion with 100-fs resolution. The decay time of the $n=2$ electron-to-heavy-hole transition $(e,\mathrm{hh})_2$ depends both on the excess energy of the charge carriers and on the excitation density. A Monte Carlo simulation allows us to reproduce the experimental data with high accuracy. The intrinsic LO-phonon scattering rate is found to be 2.0×10^{12} s^{-1} for 80-meV subband separation. We show the wave-vector dependence and explain the density and excess energy dependence. [S0163-1829(96)52340-4]

In quantum wells, LO-phonon emission is the most important energy relaxation channel for electrons. The probability of interaction is proportional to $1/|q|^2$ (q is the wave vector of the emitted phonon),[1] and to the overlap of the wave functions of the initial and final electron states.[2] If the intersubband separation is larger than the LO-phonon energy, the intersubband scattering is expected to be subpicosecond.[1] If, on the contrary, the subband separation is smaller than 36 meV, the scattering time is due to acoustical phonons and carrier-carrier (CC) scattering. In this case and for carrier densities $<10^{11}$ cm^{-2} the decay rate should be greatly reduced.[3]

Femtosecond dynamics in quantum wells have been investigated using different techniques: Raman scattering, pump-probe, and photoluminescence (PL) up-conversion.[3–5] A coherent picture emerges for the cooling and capture rates.[6–8] For intersubband scattering, if a consensus now exists on the theoretical description of phonons,[9,10] the experimental situation is not clear. Different techniques have delivered different values: this is true both for the case of an intersubband spacing smaller than 36 meV (Refs. 4, 11, and 12) and when it is larger than 36 meV.[13–16]

A resurgence of the interest in this subject has developed recently due to the demonstrated feasibility of infrared intersubband quantum-well lasers.[17] An exact knowledge of the intersubband scattering processes is fundamental in order to understand how these lasers work. Our results are obtained by PL up-conversion, a technique which allows the direct and sensitive determination of the dynamics of the photogenerated charge carriers in time- and energy-resolved measurements down to very low densities, keeping a 100-fs time resolution. In this paper, we will show how the results of the experiment may be obscured by different mechanisms (depending on the excitation density and on the photon energy), and we will give final results that are in very good agreement with a Monte Carlo simulation of the experiment including all the relevant scattering processes.

A resolution of 100 fs is obtained by up-conversion in a $LiIO_3$ crystal. Two different laser sources have been used, a synchronously pumped dye laser at 600 nm and a Ti:sapphire laser at 705 nm. Multiple quantum-well samples (25 wells) with different well widths have been studied, all with 70-Å barriers of either AlAs or $Al_{0.45}Ga_{0.65}As$. Our samples were grown by molecular beam epitaxy and the sample parameters were checked by a combination of PLE measurements and x-ray-diffraction studies. We concentrate on the results from similar samples, with well widths between 90 and 135 Å. The energy spacing between the first and the second confined electron levels is about 100 meV. Differences in confinement energy arise from the changes in the Al concentration, as well as in the width of the well. The excitation density was varied between 5×10^{10} cm^{-2} and 10^{12} cm^{-2} and the lattice temperature was kept at 77 K, allowing a reasonable thermal population in the second heavy-hole subband.

Figure 1 shows energy resolved spectra of a 135-Å (92-Å) well sample, with an $Al_xGa_{1-x}As$ (AlAs) barrier, under different excitation conditions (density and wavelength). The spectra are dominated by the $(e,\mathrm{hh})_1$ transition at about 1.55 eV. Since the PL intensity is proportional to the joint density of states and to the product of the Fermi distribution functions of electrons and holes, the spectra yield a variety of information concerning the carrier thermalization and the degree of thermal equilibrium between the different subbands. At this point it is important to emphasize that all dynamical processes concerning the hole population in the QW occur beyond the resolution of our experiments. The effective hole thermalization occurs on a time scale less than 50 fs, due to the small excess energy of the holes and the effectiveness of hole-hole scattering.[18] As the holes have a small excess energy and as we keep the lattice temperature above 50 K, their cooling has a negligible influence on the spectra.

The electron temperature manifests itself as an exponential slope on the high-energy side of the luminescence.[19] If the electrons in subbands e_1 and e_2 are in thermal equilibrium (i.e., same chemical potential), the PL spectra should show a steplike increase at the onset of the $(e,\mathrm{hh})_2$ transition by a factor of 2 corresponding to the underlying change in the density of states. Such a situation is observed in the upper spectrum of Fig. 1(a). On the contrary, nonequilibrium is evidenced in other spectra displayed in Fig. 1. The lower

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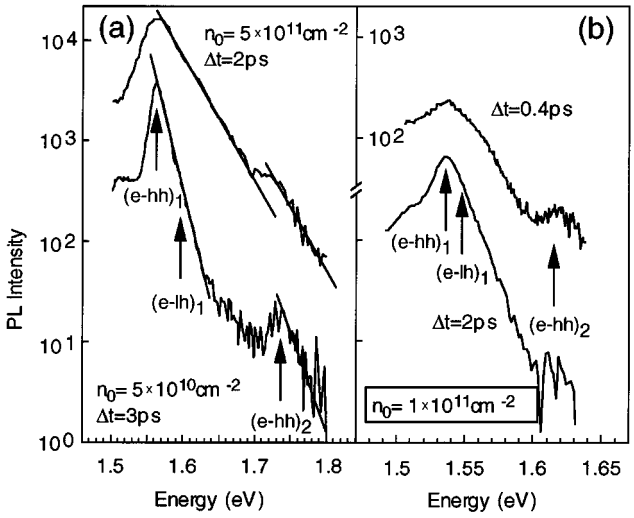

FIG. 1. Energy resolved spectra of GaAs QW samples with 175 meV (AlAs barrier) (a) and 80 meV ($Al_xGa_{1-x}As$ barrier) (b) subband spacing. The well widths are 92 and 135 Å, respectively. (a) Depending on the excitation density, the spectra show a step increase in intensity, with a factor varying from 2 to 100, at the $(e,\mathrm{hh})_2$ transition energy. In the case of a low excitation density the carrier temperatures in the first and second subband are not the same. (b) Spectra at 0.4 and 2 ps after the excitation. The nonexponential shape of the high energy side of the $(e,\mathrm{hh})_1$ transition in the upper spectrum shows that the carriers are not fully thermalized at times shorter than 400 fs.

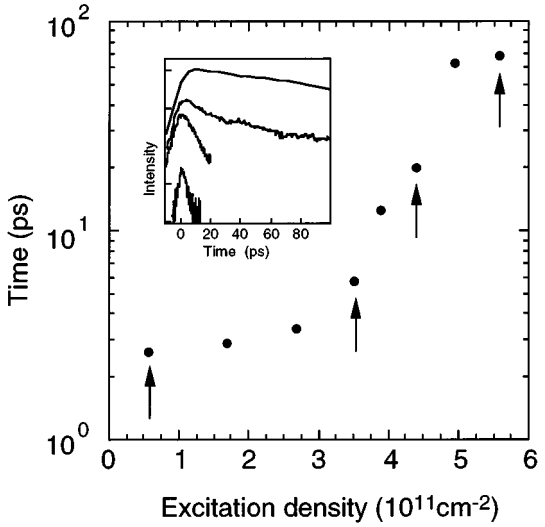

FIG. 2. The decay times of the $(e,\mathrm{hh})_2$ transition show a strong dependence on the excitation density. The results were obtained with a 92 Å GaAs/AlAs sample and 600-nm excitation. The inset presents a selection of transients (see arrows).

trace of Fig. 1(a) shows a higher temperature in e_2 than in e_1. In addition, the PL intensity jumps by a factor of 100 at the onset of the $(e,\mathrm{hh})_2$ transition indicating clearly that e_1 and e_2 are not in thermal equilibrium.

Note that in the short time spectra (lower curve, left panel and upper curve, right panel), the high energy tail of the luminescence appears to be not strictly exponential, which might be an indication for a nonthermalized carrier distribution.

It is important to realize that the intersubband scattering rates can only be determined from a starting situation with different chemical potentials. Only in that case is the PL decay at the $(e,\mathrm{hh})_2$ transition dominated by electron transitions into the first subband equilibrating the distribution functions related to each subband. Otherwise, the PL decay time corresponds to simultaneous carrier cooling in both subbands. Nevertheless, even if nonequilibrium exists between the two subbands at early times, the intersubband scattering time can be masked by competing mechanisms. We will therefore show in the following that it can only be directly identified in the decay spectra if (a) the excitation energy is chosen such that no states higher than the e_2 level are excited, and (b) the excitation density is $\leqslant 3\times10^{11}$ cm^{-2}.

We plot in Fig. 2 the variation of the decay time of the $n=2$ luminescence in one of the samples, under excitation at 600 nm. At low enough densities, the relaxation time has a value of 3 ps. This limit does not depend on the well thickness.[20] The observed increase of the decay time for increasing excitation densities shown in this figure is clearly an effect of Fermi filling of the final states. This filling goes together with both an increase of the electron-electron scattering rate, which allows faster intrasubband and intersubband thermalization, and with the creation of a hot-phonon population. As a result, backscattering of carriers from $n=1$ to $n=2$ via phonon reabsorption becomes more and more possible. This process is favored in thicker wells where the energy spacing is smaller, and we indeed observe that the density required for longer decay times gets smaller and smaller as the well width is increased.

When smaller excitation energies are used, the same density-dependent behavior is observed but the low density limit of the time decay is much shorter (see Fig. 3). Depending on their excess energy, a portion of the carriers can indeed be scattered very fast into satellite valleys. At 2.06 eV the carriers are generated above the L valley in GaAs (1.82 eV) and can scatter within 100 fs. They need a longer time delay to return to the Γ valley.[21] The photoluminescence decay of 3 ps, obtained for low carrier densities and excitation energies above the L valley, is then not given by the intersubband scattering rate but by the intervalley scattering. A simple rate equation model can show that in a three-level system, with two characteristic times for transitions into and from the intermediate level, the shorter time always occurs as the rise time and the longer one as the decay time. Thus the rise time of the luminescence corresponds basically to the intersubband scattering time ($\approx$600 fs), and the decay of the curve to the arrival time of the carriers on $n=2$: the intervalley scattering time. The value of 3 ps is in good agreement with experiments on bulk GaAs.[20]

In Fig. 3 are displayed the decay curves for the case of excitation at 705 nm (in the $n=3$ subband for a 135-Å sample with $Al_{0.45}Ga_{0.55}As$ barriers and in the $n=2$ subband for a 135 Å sample with AlAs barriers). We observe much shorter decay times than when the sample is excited at 600 nm. The reason is clearly that the photoexcited carriers can only scatter to lower-lying confined subbands, and not to satellite valleys. From the slope of the curves we extract decay times of 800 fs and 1 ps. However, these times cannot be considered as the real intersubband scattering times due to the complex relaxation processes occurring in the experiment. In particular, at an excitation in the third subband,

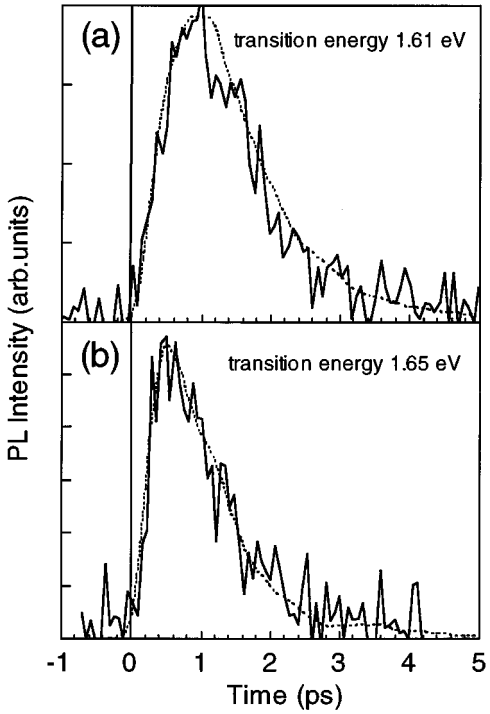

FIG. 3. Full line, decay curves of the $n=2$ luminescence for two 135-Å MQW samples with either AlAs or $Al_xGa_{1-x}As$ barriers. Dashed line results of the Monte Carlo simulation. In (a), the third subband is excited, in (b) only the first and second subband are excited.

electrons coming from the $n=3$ subband feed the $n=2$ subband with some delay (see Ref. 22).

Even in the simplest case (injection only in the $n=2$ subband), the complexity of the initial photoexcited state (several populated valence and conduction subbands), the importance of carrier-carrier (CC) scattering, and hot-phonon reabsorption led us to the use of an ensemble multisubband Monte Carlo simulation. Our simulator includes conduction bands, heavy- and light-hole bands, but neglects higher conduction bands and the spin-split-off valence band, which are not excited in the experiments. Electron and hole wave functions have been derived from an envelope function calculation within the effective-mass approximation. LO confined modes and interface modes are included within the dielectric continuum model.[23] The electron intersubband scattering times predicted by this model are 500 f and 630 fs, respectively, for the two quantum wells. Although the carrier density is relatively low, we have included CC scattering. Both intersubband and intrasubband CC scattering are considered within the static screening approximation: the multiband, multisubband dielectric function $\varepsilon(\omega,q)$ is computed for $\omega=0$, but including the full wave-vector dependence, and it is updated every 50 fs to take into account the evolution and change in the carrier distribution function. For low density conditions the static screening approach gives almost the same result as a full dynamical screening.[24] Finally, interband and intraband hole scattering by bulk TO phonons is also included. For the calculation of the luminescence signal the carrier distribution function is convoluted with the temporal profile of the laser exactly as it happens in the upconversion experiment.

In Fig. 3 we can see how the Monte Carlo simulations (dashed line) fit perfectly the decay curves. Our results confirm that the intersubband relaxation times indeed depend on the phonon wave vector. For the two samples of Fig. 3 the wave vectors involved are 2.7×10^8 m^{-1} and 3.3×10^8 m^{-1} for the 80 and 100 meV subband spacing, respectively. The dependence of the decay curve on excitation density is also well reproduced. A slow down of the decay curve is interpreted as being due to an increasing influence of Fermi filling, hot phonons, and carrier-carrier scattering. It is important to notice that the excellent theoretical fit presented in this paper has been obtained with exactly the same Monte Carlo simulator used in Ref. 13. This is a further strong proof of the validity of the theory and of the extreme sensitivity of both experiments.

Some recent experimental results do not agree with the times obtained above.[14,16] We think that the difference may be due either to high density effects, importance of satellite valleys, or excitonic influence, all effects that have been carefully avoided in the present experiment.

In conclusion, with our joint experimental and theoretical investigation we were able to measure the intersubband scattering time in several $GaAs/Al_xGa_{1-x}As$ quantum wells. Our luminescence results are in excellent agreement with the theory and with previous transmission experiments on similar samples. We show, using the same experimental conditions for more than one sample, that for a subband splitting of about 100 meV the intersubband scattering rate is 1.6×10^{12} s^{-1}, and we clearly show its dependence on the q vector of the emitted phonons. The use of differing excitation densities and energies gives more insight in the processes depending on these parameters (L-valley scattering, hot-phonon effects) and enables us to fully understand the difference with previous luminescence measurements. The observed energy spectra allow us at the same time to monitor directly the density and interaction conditions in the subbands. From these spectra we obtain information about nonthermalization effects of the carriers within the first hundreds of femtoseconds and about weak CC interaction between the subbands at low excitation densities, information necessary for the exact knowledge of our experimental conditions.

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