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Femtosecond intersubband relaxation in GaAs quantum wells

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We investigate the intersubband relaxation in GaAs quantum wells at room temperature using differential transmission spectroscopy with subpicosecond time resolution. The dynamics of the subband populations are derived from the experimentally observed reduction of oscillator strength of the corresponding exciton transitions. The obtained electron intersubband scattering times are 160 fs for relaxation from the second to the first subband in a 15-nm quantum well and 170 fs for relaxation from the third to the first subband in a 20-nm structure. These times are significantly shorter than those deduced from previous experiments and from some theoretical studies.

In recent years, a large number of experimental and theoretical studies have been devoted to carrier relaxation in semiconductors. Time-resolved optical methods have been applied to study directly the thermalization and cooling of carriers in bulk semiconductors and quantum wells (QW's) (for an overview, see, e.g., Refs. 1 and 2). One topic of particular interest has been the investigation of specific differences between carrier relaxation in bulk and quantum well structures. It has been found that relaxation processes generally occur on the same time scale in bulk semiconductors and QW's. However, in the case of quantum wells, one has to distinguish between carrier cooling within one subband (intrasubband relaxation) and scattering of carriers from higher subbands into the lowest subband (intersubband relaxation), which is a process not present in the bulk.

Various experimental techniques, such as time-resolved Raman scattering,[3,4] pump-probe,[5–8] and photoluminescence measurements[9,12] have been applied to determine intersubband relaxation rates. It is generally accepted that the intersubband relaxation in GaAs quantum wells occurs primarily by LO-phonon emission when the subband splitting is larger than the LO-phonon energy $\hbar\omega_{\mathrm{LO}}$,[3,13] and when the carrier density is below $\approx 10^{12}$ cm^{-2}.[14,15] The intraband LO-phonon emission time is about 170 fs in the bulk and similar in QW's.[16,17] For subband splittings smaller than $\hbar\omega_{\mathrm{LO}}$, carriers at the higher subband edge can no longer emit LO phonons and the intersubband relaxation becomes rather slow. For this situation, relaxation times on the order of several hundred picoseconds for low carrier density[3,12] and 20–40 ps for high carrier density[8] have been reported. A number of experiments have shown that the intersubband relaxation is indeed much faster when the subband splitting is larger than the LO-phonon energy.[3,4,7,8] However, due to the limited time resolution of all of these experiments only an upper limit could be given for the intersubband relaxation rate τ_{21} which describes the scattering of electrons from the second into the first subband. The lowest value of $\tau_{21} < 1$ ps is estimated in Ref. 4. A value for τ_{21} of 600–650 fs has been derived indirectly from photoluminescence measurements in biased superlattices[9] and asymmetric coupled QW's.[10] In doped, narrrow GaAs QW's, Faist *et al.* have determined a relaxation time of 650 fs from the temperature dependence of the intersubband absorption.[11]

In modulation-doped QW's much slower (by an order of magnitude) intersubband relaxation has been measured.[5] Initially, this observation has been interpreted as evidence for a strong influence of confined phonon modes.[18] Subsequent studies have shown, however, that the slow relaxation is caused by scattering into barrier states and intervalley scattering.[6,19]

Several theoretical studies of carrier relaxation predict quite different scattering rates for intrasubband and intersubband processes.[13,20–22] Although both processes occur mainly by LO-phonon emission, different scattering rates may arise from different k-selection rules. Several models have been proposed to account for confined phonon modes, and the reported scattering rates have been found to depend strongly upon the chosen model.[20–23] The calculated values of the interband scattering rate τ_{21} in a 15-nm GaAs QW vary from ≈ 250 fs (Ref. 13) to 2 ps.[21] Some of the earlier calculations are critically reviewed in Ref. 24, where it is argued that the scattering rates are independent of the type of phonon modes considered, as long as complete sets of vibrational wave functions are used.

A precise knowledge of intersubband relaxation rates is not only important for a comparison with theoretical approaches, but also for application purposes: Intersubband relaxation may be a limiting factor for the high-speed performance of sophisticated semiconductor devices like resonant tunneling transistors, far-infrared photodetectors, or quantum well lasers.[25]

In this paper, we present our experimental data of intersubband relaxation obtained with subpicosecond time resolution. We address the most fundamental situation of undoped, uncoupled, and unbiased QW's. Carriers are optically excited in the GaAs/Al$_x$Ga$_{1-x}$As QW's at room temperature, i.e., in the temperature regime of useful device operation. We investigate relaxation rates for subband splittings larger than the LO-phonon energy $\hbar\omega_{\mathrm{LO}} = 36$ meV both for relaxation from the second and third electronic subband. As experimen-

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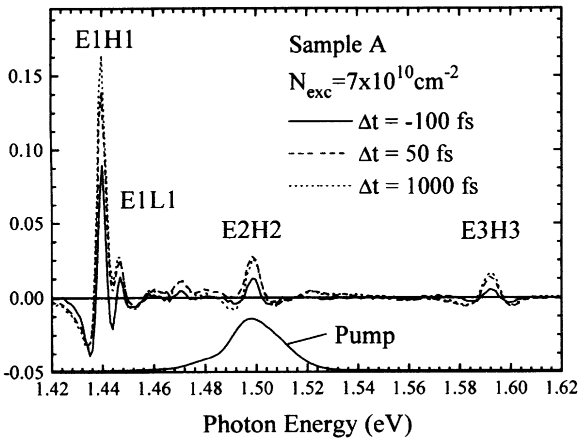

FIG. 1. Time-resolved diffferential transmission spectra (DTS) of sample A at time delays of -100 fs, 50 fs, and 1 ps. The peaks correspond to exciton transitions in the linear absorption, which are bleached by the pump-induced carriers.

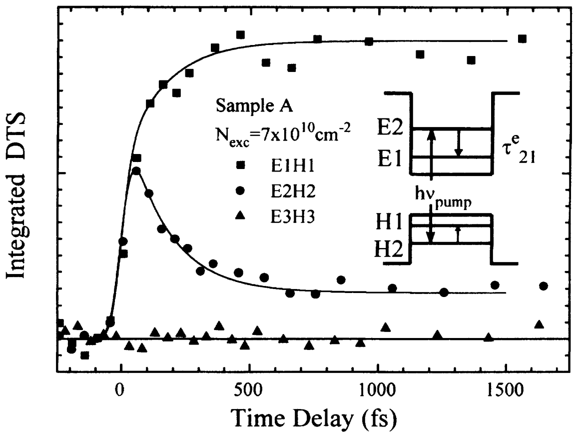

FIG. 2. Time evolution of the integrated absorption change at the $E1H1$, $E2H2$, and $E3H3$ exciton in sample A for the same experimental condition as in Fig. 1. The lines are simulations obtained from coupled rate equations describing the subband occupations.

tal technique, we use time-resolved measurements of exciton bleaching at different subband edges to monitor the time evolution of the respective subband occupations. The experiments are performed in a standard pump-probe setup, using a white-light continuum probe and near infrared (NIR) pump pulses of 80–90-fs duration. The white-light continuum is obtained by focusing amplified femtosecond pulses of a colliding pulse mode-locked dye laser into an ethylene glycol jet. A part of this continuum is spectrally filtered and amplified in a NIR dye (Styryl 9m) amplifier stage to obtain pump pulses that are tunable between 1.44 eV and 1.57 eV. Both amplifier stages are pumped by the same 6.8-kHz copper vapor laser. We use an optical multichannel analyzer system to measure differential transmission spectra (DTS), i.e., pump-induced transmission changes $\Delta T/T_0$, as a function of the delay between pump and probe. The DTS approximately give the negative change of the optical density, $-\Delta\alpha d$, when this change is small, and when reflectivity changes are neglected. The intrinsic chirp of the probe continuum which corresponds to a frequency-dependent time delay is taken into account in the data analysis.

The samples are multiple quantum wells with 40 periods of GaAs wells and 20-nm $Al_{0.35}Ga_{0.65}As$ barriers grown by molecular beam epitaxy. The nominal well widths are 15 nm (sample A) and 20 nm (sample B). The GaAs substrate is removed over an area of $\approx 1\times 1$ mm^2 by selective etching to allow transmission measurements. Excitonic absorption peaks are clearly resolved in the linear absorption and in the photoluminescence excitation spectra of the samples. The positions of these peaks agree well with calculated values. The calculated subband splitting between the first and second electronic subband, $E1$ and $E2$, is 50 meV in sample A and 31 meV in sample B, i.e., significantly larger and smaller than $\hbar\omega_{LO}$, respectively. The splitting between the first and the third electron subband in sample B is 82 meV. All corresponding heavy-hole subband splittings are much smaller than the LO-phonon energy.

Figure 1 shows typical DTS of sample A at different time delays. The pump wavelength has been tuned to the excitonic transition between the second heavy hole and the second electron subband, $E2H2$. As a result, electrons and holes are created at the minimum (maximum) of the second electron (heavy-hole) subband and in continuum states of the lowest subbands. From the linear absorption we estimate that about 50% of the carriers are excited into the second subbands. The total carrier density is 7×10^{10} cm^{-2}, as estimated from the pump power and spot size. The DTS show absorption saturation, i.e., bleaching, of the excitons at the $E1H1$, $E1L1$, $E2H2$, and $E3H3$ transitions, where H denotes the heavy-hole and L the light-hole transitions. The spectra at different delay times indicate a rather slight variation of the exciton line shape and absorption strength after the pump pulse has passed. A very striking fact is the saturation of the $E3H3$ transition, since no carriers are excited into the third subbands. Obviously, the DTS are dominated by excitonic effects for this excitation density and therefore do not reflect the free carrier distribution functions (via bleaching of continuum transitions), as in bulk GaAs.[2,26]

Different physical processes contribute to the exciton bleaching. Phase-space filling (PSF), i.e., the occupation of single-particle states which contribute to the exciton wave function, causes a reduction of the exciton oscillator strength. Coulomb screening reduces the electron-hole interaction and leads mainly to a broadening of the exciton lines. The latter effect is not directly related to the occupation of specific carrier states and affects also excitons related to unoccupied subbands. From a phenomenological point of view, a reduction of an excitonic absorption peak may result from a reduction of the oscillator strength (ROS) of the exciton, from a shift of the transition energy, or from a broadening of the exciton line. From DTS measurements, the ROS contribution can be separated by integrating the absorption changes over the spectral width of the excitonic transition. In a previous paper, we have shown that this integrated signal is approximately proportional to the carrier density and is not influenced by intrasubband carrier cooling.[27] Therefore, it can be used as a *direct measure of the subband population.*

In Fig. 2, we present the temporal evolution of the ROS via integrated DTS of the $E1H1$, $E2H2$, and $E3H3$ excitons for the same experimental conditions as shown in Fig. 1. The values obtained for the $E1H1$ also contain the small contribution from the $E1L1$, because of the small energy difference between these two transitions. This should be of minor importance, since we will show that the dynamics is mainly determined by the electrons. The absorption strength of the $E3H3$ exciton remains unchanged at all delays, showing that the corresponding subbands remain unoccupied. However, the ROS of the two lowest excitons differ significantly in their time dependence: Initially both signals rise with the integral of the pump pulse (i.e., without delay). The ROS of the $E2H2$ transition, however, exhibits a fast decay during the first 500 fs, while the ROS of the $E1H1$ transition displays a complementary delayed rise in the same time interval. After 1 ps, both signals reach quasiequilibrium values, and do not change further on a picosecond time scale. The maximum ROS of the $E2H2$ exciton is about 50% of the ROS maximum at the $E1H1$ transition. These data obviously suggest that there is a fast intersubband relaxation from the second into the first subband. In principle, this might occur for electrons ($E2 \rightarrow E1$) and for holes ($H2 \rightarrow H1$). However, the observation that the ROS of the $E2H2$ exciton does not decay to zero, but reaches a quasiequilibrium value, indicates that a fast intersubband relaxation occurs only for one type of carriers. We do not observe an ROS of the $E2H2$ exciton when the sample is excited below this transition. As the hole subband splitting is only 11 meV, fast intersubband scattering is only expected for the electrons. In fact, Höpfel *et al.*[12] have determined an extremely long $H2 \rightarrow H1$ relaxation time of 130 ps (at low temperature) for a sample with similar parameters as the one used in our experiment. Therefore, it is most plausible to interpret the data in Fig. 2 in terms of fast electron intersubband relaxation. This is also consistent with the observation that the quasiequilibrium ROS at $E2H2$ is much lower than the maximum value. If one assumes that the ROS is mainly caused by phase-space filling, this result would indicate that PSF by holes is less effective than by electrons of the same density. This can be expected, since the hole occupation numbers f_h will be much lower than the electron occupation f_e due to the much larger density of states in the valence band.

We have simulated the experimental data by a set of coupled rate equations for electron and hole occupations of the first and second subbands, n_{E1}, n_{H1}, n_{E2}, n_{H2}. We assume a generation of carriers in all four bands according to a Gaussian pump pulse of 80-fs duration, a fast electron relaxation with a time constant τ_{21}^{e}, and a very slow hole relaxation time $\tau_{21}^{h}=50$ ps (the results obviously do not depend on the precise value). Finally, the ROS for each exciton is taken as the sum of an electron and a hole contribution, i.e., $c_E n_E + c_H n_H$, where c_E and c_H define the efficiency of ROS by electrons and holes, respectively. We assume the same ratio c_H/c_E for the first and second subband. The time constant τ_{21}^{e} and the ratio c_H/c_E are determined by fitting the experimental data. The best fit of the measurement is shown by the lines in Fig. 2. We find a relative bleaching efficiency of holes $c_H/c_E=0.25$ and a time constant $\tau_{21}^{e}=160$ fs.

An analogous experiment with sample B, where the $E2$-$E1$ splitting is less than $\hbar\omega_{\mathrm{LO}}$ does not show the characteristic time dependence as observed in Fig. 2. This is illustrated in Fig. 3, where we present the time dependence of the ROS of the lowest three electron–heavy-hole excitons in sample B after excitation resonant with the $E2H2$ transition. Apart from correlated variations of the $E1H1$ and $E2H2$ signals, which we attribute to pump intensity fluctuations, there seems to be a very slight enhancement of the $E2H2$-ROS around time delay zero. Most probably, this corresponds to a background bleaching by carriers which are excited into the continuum of the lowest subbands and which thermalize within less than 200 fs.[2] However, the data show neither the strong decrease of the $E2H2$ signal nor the complementary increase of the $E1H1$ bleaching observed in sample A. There is no significant change of the signals within 40 ps indicating a much slower intersubband relaxation due to the low splitting between the $E2$ and $E1$ subbands.

However, the energy differences between the third and the lowest two electron subbands in sample B are larger than the LO-phonon energy, so that again a fast intersubband scatter-

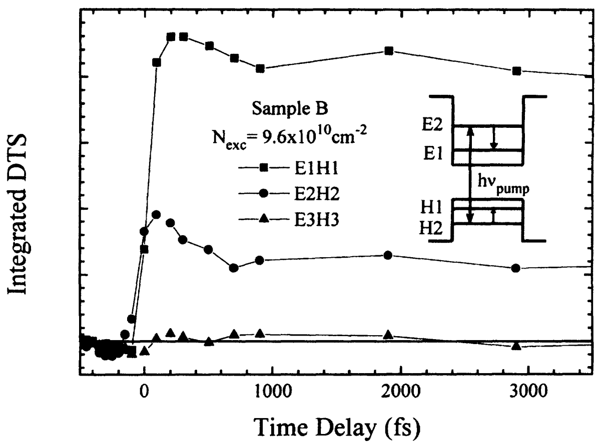

FIG. 3. Time evolution of the integrated absorption change at the $E1H1$, $E2H2$, and $E3H3$ exciton in sample B, after excitation resonant with the $E2H2$ transition.

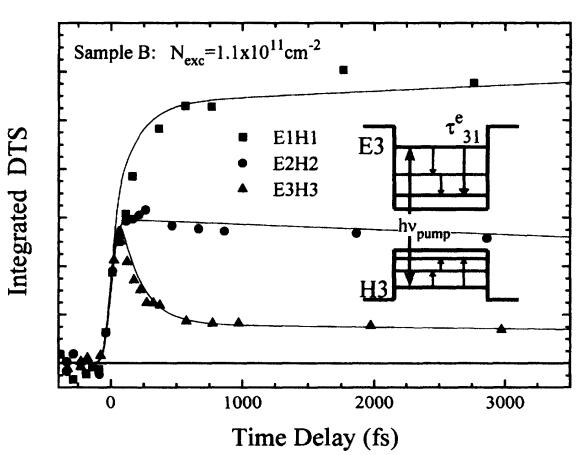

FIG. 4. Time evolution of the integrated absorption change at the $E1H1$, $E2H2$, and $E3H3$ exciton in sample B, after excitation resonant with the $E3H3$ transition. The lines are calculated with coupled rate equations describing the subband occupations.

ing can be expected. This has been investigated by exciting sample B at the $E3H3$ transition. In Fig. 4, we show the time dependence of the ROS of the $E1H1$ to $E3H3$ transitions. The ROS of the $E4H4$ line remains zero in this experiment. The excitation density was 1.1×10^{11} cm^{-2} for this experiment. We find a subpicosecond decay for the ROS of the $E3H3$ and again a corresponding increase at the $E1H1$ transition, while the bleaching of the $E2H2$ only shows a very slow decay. This indicates that a fast electron relaxation occurs predominantly from the third into the first subband. Again, the time dependence can be well fitted by a set of rate equations, now taking into account six electron and hole bands. This is shown by the lines in Fig. 3. The best fit of the data has been obtained assuming a relaxation time constant $\tau_{31}^{e}=170$ fs. The slower decay of the $E2H2$ signal has been modeled by a $E2\rightarrow E1$ relaxation with a 20-ps time constant. Since *hot* electrons are injected into the $E2$ band in this experiment (in contrast with pumping at the $E2H2$ transition), electron relaxation from the second into the first subband may occur through LO-phonon emission by carriers in the high energy tail of the energy distribution.

In conclusion, we present an experimental investigation of intersubband relaxation in GaAs quantum wells with subband separation larger and smaller than $\hbar\omega_{\mathrm{LO}}$. From time- and spectrally resolved measurements, we determine the dynamics of the subband occupation with subpicosecond time resolution. Our data clearly show a fast electron intersubband relaxation time τ_{21}^{e} of 160 fs for quantum wells with 15-nm well width and a relaxation time τ_{31}^{e} of 170 fs for wells with 20-nm width. The first value is close to the value calculated by Ferreira and Bastard.[13] Both values are very close to the LO-phonon emission time in bulk GaAs, suggesting that the layered structure of the QW's has only a negligible influence on the net electron–LO-phonon scattering rate, at least for the present experimental conditions (room temperature and large well widths). This observation agrees with studies of carrier cooling, which have obtained similar relaxation rates in bulk material and quantum wells.

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