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Relaxation and dephasing of the intersubband transitions in \textit{n}-type InAs/AlSb multi quantum wells

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Intersubband electron relaxation and dephasing has been studied in InAs/AlSb multi quantum wells using time resolved femtosecond spectroscopy. The authors have measured a relaxation time $T_1$ of 3 ps at $T=300 \text{ K}$ (and 4.6 ps at $T=10 \text{ K}$) for a transition energy of 260 meV, which is in good agreement with our calculations. A dephasing time $T_2$ of $\sim 320 \text{ fs}$ for optically excited electrons in the second subband was measured which determines the homogeneous broadening ($\sim 4.1 \text{ meV}$) of the absorption line. © 2007 American Institute of Physics. [DOI: 10.1063/1.2769948]

The InAs/AlSb heterostructure has attracted considerable interest since it provides unique physical properties. The low effective electron mass in InAs provides excellent mobility, long electron relaxation time, and a high intersubband optical matrix element, while the high conduction band of low effective electron mass in InAs provides excellent mobility. These favorable properties have led to the adoption of this material system for the realization of high-performance quantum cascade lasers (QCLs) at the short wavelength limit of the midinfrared spectrum. Laser emission from InAs/AlSb QCLs has been demonstrated at wavelengths as short as 2.95 $\mu$m, while spontaneous intersubband emission from QC light emitting diodes has been observed down to 2.5 $\mu$m.\textsuperscript{1,2}

One of the key parameters for QCL design in any material system is the nonradiative intersubband relaxation time, since this determines the intersubband population inversion. However, despite its potential importance for QCL technology, there have been no previous reports on intersubband electron relaxation in the InAs/AlSb system.

In this letter, we describe results of femtosecond pump-probe transmission measurements on an InAs/AlSb multi quantum well sample which allow us to determine the intersubband relaxation time ($T_1$) directly. We compare the obtained results with calculations assuming longitudinal optical (LO) phonon scattering to be the dominant relaxation mechanism. Additionally, we perform linewidth analysis of the intersubband transition (ISBT) by comparing results from a femtosecond four wave mixing experiment, where we measured the dephasing time $T_2$, with line-shape fitting and found that the ISBT absorption spectrum is dominantly inhomogeneously broadened.

The InAs/AlSb multi quantum well sample studied in this work was grown by molecular beam epitaxy. Because no semi-insulating lattice matched substrate (e.g., InAs) is available for InAs/AlSb heterostructures, a semi-insulating GaAs substrate was used and a 1.5 $\mu$m thick, undoped AlSb buffer layer was grown on it to accommodate the lattice mismatch.\textsuperscript{3} After the buffer layer, 30 periods of AlSb (15 nm)/InAs (7.5 nm) were grown. The InAs quantum wells were doped with tellurium to about $2 \times 10^{12} \text{ cm}^{-2}$. The structure was capped with a 10 nm thick InAs layer to prevent oxidation of the last AlSb layer.

Figure 1 shows the linear absorption spectra at $T=10 \text{ K}$ and $T=300 \text{ K}$ obtained using Fourier transform infrared (FTIR) spectroscopy. The displayed absorption curves are the ratio between the spectra obtained with light polarized in growth direction (TM polarized) and light polarized perpendicularly to the growth direction (TE polarized). Since only the TM polarization couples to the ISBTs, the TE polarization spectra can be used as a reference. A prismlike geometry\textsuperscript{4} with one pass through the quantum well layers was used to provide the intersubband absorption, as shown in the inset of Fig. 1. A strong intersubband absorption peak at 262±2 meV (~4.8 $\mu$m) was observed at $T=10 \text{ K}$, which is redshifted by ~7 meV at room temperature. The full width at half maximum of the absorption peak is ~45 meV at $T=10 \text{ K}$, increasing to ~50 meV at room temperature. These results are in agreement with observed ISBT for similar structures reported in Ref. 5.

Midinfrared, femtosecond laser pulses from an optical parametric amplifier (1 kHz repetition rate, 110 fs pulse width) were used to perform the pump-probe and four wave

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Intersubband absorption spectra from an InAs/AlSb multi quantum well sample at $T=10 \text{ K}$ and $T=300 \text{ K}$. The inset shows the sample geometry used in order to obtain intersubband coupling.}
\end{figure}
mixing measurements. A fast, photovoltaic HgCdTe detector in combination with a boxcar integrator was used to detect the signal.

A typical pump-probe signal at \( T = 10 \, \text{K} \) and \( \lambda = 4.8 \, \mu\text{m} \) is shown in Fig. 2. The applied pump power density was 0.2 MW/cm\(^2\) and the probe pulses were attenuated to be about 200 times weaker than the pump pulses using neutral density filters. A relaxation time of \( T_1 \sim 4.6 \, \text{ps} \) can be extracted by applying a single exponential fit to the pump-probe curve. The temperature dependence of the relaxation time is shown in the inset of Fig. 2. A decrease with higher \( T \) is shown in the inset of Fig. 2. A decrease with higher \( T \) is observed and the dependence agrees well with the fit for LO phonon emission according to \( \tau^{-1} = \Gamma_0 [N_{\text{LO}}(T) + 1] \), where \( \Gamma_0 \) is the scattering rate at \( T = 0 \, \text{K} \) and \( N_{\text{LO}}(T) = [\exp(E_{\text{LO}}/k_B T) - 1]^{-1} \) is the Bose-Einstein distribution function for LO phonons.

In order to compare the experimental results with the expected values for the intersubband relaxation time, we have performed calculations similar to those presented in Ref. 2. The electron effective mass \( m^* \) for InAs quantum wells and the conduction band nonparabolicity coefficient \( \gamma \) (defined as in Ref. 6) were used as fitting parameters. It has been shown, for example, in Ref. 2 that the effective mass for InAs/AlSb quantum wells can be slightly different from the bulk value of 0.023\( m_0 \) depending on the substrate the structures have been grown on (InAs or GaSb). For the sample studied in this work (grown on GaAs substrate with thick AlSb buffer layer), the final structure can be slightly strained, consequently leading to the changes in the electron effective mass. Performing the calculations, we have found that an effective mass value \( m^* = 0.019 m_0 \) and a nonparabolicity parameter \( \gamma = 4.8 \times 10^{-18} \, \text{m}^2 \) fit best with both the energy of ISBTs and the intersubband relaxation time. These values are very similar to those presented in Ref. 2 for the InAs/AlSb quantum wells grown on GaSb. The relaxation time \( \tau \) calculated using the above set of parameters and LO phonon scattering as the main scattering mechanism is 2.5\( \pm \)0.25 \( \text{ps} \) at 300 \( \text{K} \) and is slightly shorter than the measured time of 3\( \pm \)0.3 \( \text{ps} \). Apart from a tolerance arising from experimental errors and uncertainties in calculation input parameters, the difference in the values could arise from the fact that only intersubband but not intrasubband scattering has been included in the calculations. It has been shown in Ref. 8 that for high intersubband energies and strong nonparabolicity (like in InAs), intrasubband scattering also needs to be taken into account.

Comparing the intersubband relaxation times of 3 ps at \( T = 300 \, \text{K} \) and 4.6 ps at \( T = 10 \, \text{K} \) with other III-V material system\(^{8-12} \) shows that they are relatively long, taking the transition energy into account. This is theoretically predicted because of the very low effective mass of InAs. A further reason for the long relaxation time is the low InAs longitudinal optical phonon energy (30 meV), leading to a larger in-plane wave vector that needs to be exchanged between phonon and electron for scattering between subbands.

After measuring the relaxation time using pump-probe spectroscopy, we performed degenerate time-integrated four wave mixing (TI-FWM) experiments\(^{13,14} \) on our sample. These experiments are carried out to determine the excited state electron dephasing time which is directly related to the homogeneous linewidth of the absorption peak.\(^{13} \) Figure 3 shows a typical signal of the TI-FWM intensity over the time delay between the first and the second pulse. A time constant \( \tau_{\text{FWM}} \) of \( \sim 80 \pm 5 \, \text{fs} \) is extracted from the measurement. Although the FWM decay time is found to be shorter compared to the MIR pulse width, we were able to extract this value very accurately by fitting the experimental data using the convolution of two Gaussian pulses. Such a fast FWM decay time is much shorter compared to the measured intersubband relaxation time of 4.7 ps and therefore cannot be explained only by phonon-assisted electron scattering, which plays a minor role for dephasing. In Ref. 15, it was demonstrated that electron-electron scattering has a major contribution in dephasing processes for ISBT in quantum wells (QWs) even at electron concentrations as low as \( 5 \times 10^{10} \, \text{cm}^{-2} \),\(^{16} \) thus defining the homogeneous linewidth broadening.

As we will see later, the total linewidth of our ISBT absorption spectrum of \( \sim 45 \, \text{meV} \) is strongly inhomogeneously broadened, and in this case, the FWM signal decays with a time constant of \( T_2 \).\(^{15} \) Thus \( T_2 \sim 4 \tau_{\text{FWM}} = 320 \pm 20 \, \text{fs} \), resulting in a homogeneous linewidth of just \( \sim 4.1 \pm 0.3 \, \text{meV} \). Similar values have been calculated in Ref. 2 for InAs/AlSb QWs. In order to extract the inhomogeneous contribution to the total absorption linewidth, Voigt line-shape (convolution of a Gaussian with a Lorentzian) fitting was conducted with the best fit, shown in Fig. 4. Here, we fixed the homogeneous contribution (Lorentzian linewidth) of \( \Gamma_L \) = 4.1 meV, as extracted from the dephasing time measurements, and a Gaussian linewidth of \( \Gamma_G = 42 \, \text{meV} \) was found.
This $\Gamma_G$ value is very close to 44.7 meV extracted using the fit with a Gaussian peak function, thus indicating that the intraband absorption line is dominantly inhomogeneously broadened. In Ref. 17, the effects of electron-electron and electron-LO phonon scatterings on the ISBT spectra within the second Born approximation were studied for InAs QWs and very strong inhomogeneous broadening of $\sim 17.4$ meV of the absorption linewidth mainly due to distribution of transition energy (because of strong nonparabolicity in InAs QWs) was obtained. Comparing this value and our linewidth, the extra $\sim 25$ meV of inhomogeneous broadening can originate from fluctuations of the QW width, interface grading, and in our sample also from remaining strain in the structure arising from the lattice mismatch between buffer layer and substrate.

In summary, we have performed time resolved measurements on an InAs/AlSb quantum well structure and measured at an ISBT energy of 260 meV a relaxation time of $\sim 3$ ps at $T = 300$ K. Performing four wave mixing measurements, sensitive to excited carrier dephasing, we also found a dephasing time of about 320 fs dominated by electron-electron scattering. From this dephasing time, a homogeneous linewidth broadening of $\sim 4$ meV can be derived. Both these parameters play a significant role in the design and performance of quantum cascade lasers because the relaxation time influences the population inversion and the homogeneous linewidth influences the gain profile.

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