Two-photon photocurrent spectroscopy of electron intersubband relaxation and dephasing in quantum wells

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Resonantly enhanced nonlinear absorption between conduction subbands in InGaAs/AlGaAs quantum wells induces a two-photon photocurrent under femtosecond excitation, which is exploited to determine electron intersubband relaxation and dephasing times. The approach allows us to study systematically the dependence of these time constants on structural parameters, including carrier density and modulation/well doping, and to discriminate between different scattering processes. © 2007 American Institute of Physics. [DOI: 10.1063/1.2806963]

Knowing the dynamics of intersubband transitions in quantum wells (QW) is crucial for optimizing quantum well infrared photodetectors (QWIP) and quantum cascade lasers. In addition, intersubband transitions in QWs constitute a model system to study basic concepts in semiconductors, including scattering, quantum interference, and coherent transport. While most investigations have concentrated on linear spectroscopy of intersubband transitions, an increasing body of research efforts has focused on nonlinear optical studies, including harmonic generation, pump-probe, and four-wave mixing.

We have previously demonstrated two-photon photodetection involving three equidistant energy levels [\(E_2\) \(-\) \(E_1\), \(E_1\) \(-\) \(E_0\)], and quantum cascade lasers. In contrast to QWIPs, where transitions from a bound state to a continuum resonance lead to a linear photocurrent, the three-level configuration requires two photons to generate a photocurrent (inset of Fig. 2). Therefore, the photocurrent scales quadratically with the incident power, which has been verified down to excitation densities as low as 0.1 W/cm\(^2\). This quadratic behavior allows for interferometric autocorrelation measurements under femtosecond excitation, and to determine the intersubband relaxation time \(T_1\) and dephasing time \(T_2\). Based on the third-order nonlinear susceptibility \(\chi^{(3)}\), this approach provides an interesting alternative to four-wave mixing experiments for studying the dynamics of intersubband excitations.

In the present letter, we investigate systematically the influence of dopant concentration and distribution on the intersubband dynamics in InGaAs/AlGaAs QWs by interferometric two-photon photocurrent autocorrelation measurements.

The samples are based on modulation-doped and well-doped In\(_{0.53}\)Ga\(_{0.47}\)As/Al\(_{0.3}\)Ga\(_{0.7}\)As multiple QW structures grown by molecular beam epitaxy (MBE) on [100]-oriented semi-insulating GaAs substrates. The active region, designed for a transition energy \(E_2\) \(-\) \(E_1\) of about 150 meV, consists of 20 periods of 7.3 nm wide QWs separated by 46 nm wide barriers, and is embedded between \(n\)-type contact layers. For samples 1–3, nominally the central 5 nm of each QW are Si doped, whereas for sample 4, the QWs are modulation doped by incorporating 2 nm of Si-doped AlGaAs subsequent to 12 nm of undoped AlGaAs in each barrier, with doping concentrations as summarized in Table I. The wafers were processed into mesa detectors of \(120 \times 120\) and \(240 \times 240\) \(\mu\)m\(^2\) in area with Ohmic contact metallization covering the top of the mesas. For photocurrent measurements, the radiation is coupled into the structures via 45° facets in order to provide an electric field component parallel to the quantized direction. The actual doping concentrations, also given in Table I, have been measured by secondary-ion mass spectroscopy (SIMS).

Figure 1(a) shows intersubband absorption spectra of the as-grown layer structures involving the \((1) \rightarrow (2)\) transition, measured at a temperature of 77 K in Brewster angle geometry using a Fourier-transform infrared (FTIR) spectrometer. Also shown are fit functions yielding the Lorentzian full width at half maximum broadenings \(\Gamma_1\) given in Table I. The spectra in Fig. 1(a) indicate a characteristic increase of transition energy with increasing carrier density, which is attributed to many-particle effects.\(^1,4\)

### FIG. 1.
(Color online) Normalized absorption spectra (a) of the \((1) \rightarrow (2)\) transition in Brewster-angle geometry at 77 K and normalized photocurrent spectra (b) at an elevated temperature of 160 K. Solid lines in (a) indicate Lorentzian fits to the experimental spectra.

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Further information on the subband spacings is obtained from photocurrent spectroscopy at higher temperature (160 K), where photons at energy $\hbar \nu = E_2 - E_1$ produce a linear photocurrent since sufficient carriers are excited into level $|2\rangle$. After amplification by a transimpedance amplifier, photocurrent spectra are readily obtained using a standard FTIR spectrometer. Figure 1(b) shows photocurrent spectra of the well-doped structures. Interestingly, relatively narrow photocurrent peaks, only about 50% wider than those of the absorption measurement, are observed at the essentially degenerate energies of the $|1\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |3\rangle$ transitions. Besides the $|2\rangle \rightarrow |3\rangle$ bound-to-continuum excitation of electrons in the thermally populated second subband, an additional contribution presumably involves the $|1\rangle \rightarrow |2\rangle$ transition of electrons with high enough kinetic energy, such that the total energy of the final state in the second subband is close to or above the barrier edge.

Even though the $|1\rangle \rightarrow |3\rangle$ transition is parity forbidden in a symmetric QW, it still leads to a finite photocurrent, since residual asymmetry is induced by the externally applied electric field and by asymmetric dopant distributions. The steplike increase at around 280 meV indicates the photoconductive energy threshold; the broad absorption line is characteristic for bound-to-continuum transitions and relaxes the conditions for resonant two-photon transitions to be observed. Due to the induced asymmetry, the $|1\rangle \rightarrow |3\rangle$ photocurrent is larger than the peak at around 160 meV since only about 0.1% of the carriers are thermally excited into the second subband at this temperature. From Fig. 1(b), the continuum resonance appears wide enough to ensure that resonantly enhanced two-photon absorption is always present as long as the photon energy matches the $|1\rangle \rightarrow |2\rangle$ transition.

To study the intersubband dynamics, pulses of 165 fs duration, tunable from 6 to 18 μm, are generated at a repetition rate of 76 MHz by difference frequency mixing of the signal and idler beams of an optical parametric oscillator. Using a beam splitter and a Michelson interferometer, the samples are illuminated by collinear pulses with variable delay time. Measurements were conducted at a temperature of 77 K, low enough to suppress the thermally activated, linear photocurrent contribution discussed above, and at moderate operation voltages in the range of 1–2 V to avoid tunneling out of the intermediate state. The latter effect comes into play at high bias voltages where it allows for electrical switching between linear and quadratic detection.

Quadratic photocurrent autocorrelation traces, normalized to the signal at large time delay, are shown in Fig. 2. In these experiments, the excitation energy was chosen to match the $|1\rangle \rightarrow |2\rangle$ transition. The peak-to-background ratio close to the ideal 8:1 value confirms that the signal scales quadratically with the incident power for all samples. The “ideal” autocorrelation trace as obtained by assuming transform-limited Gaussian pulses (which is a good approximation for the midinfrared pulses used in our experiment) has been included in Fig. 2 for comparison.

Comparing the experimental traces with the ideal autocorrelation, two striking differences are observed. First, the fringe amplitude decays exponentially with increasing delay time $\tau$, which directly reflects the phase relaxation of the coherent intersubband polarization. Therefore, the associated decay constant agrees with the phase relaxation time $T_2$. In contrast, the ideal case (lowest panel of Fig. 2) shows a Gaussian decay of the fringes. Second, even after the fringes have disappeared, the two-photon autocorrelation signal exhibits further exponential decay towards its asymptotic value. The latter decay constant arises from the intersubband population associated with the population relaxation time $T_1$.

These considerations provide the basis for a phenomenological model introduced by Nessler et al., which yields an analytical solution for numerical fitting of the experimental data. In fact, the fits thus obtained exhibit satisfactory agreement with experimental autocorrelation traces. This agreement is also evident from the envelope functions shown in Fig. 2, which nicely reproduce the minima and maxima of the fringes.

The observed dynamics in Fig. 2, in particular, for the decay of the oscillatory part, depends considerably on the carrier density. Yet significantly longer time constants come into play for the modulation doped device structure 4. For further analysis, Fig. 3 compares the measured dynamical parameters, namely the diagonal and off-diagonal relaxation rates $T_1^{-1}$ and $T_2^{-1}$, respectively, with the decay rate $\Gamma_1/2\hbar$ associated with the $|1\rangle \rightarrow |2\rangle$ absorption linewidth. Only small deviations exist between values of $\Gamma_1/2\hbar$ and observed dephasing rates, indicating predominantly homogeneous broadening (i.e., lifetime broadening) of the $|1\rangle \rightarrow |2\rangle$ transition. Residual deviations of 10%–25% between these two quantities are attributed to some additional inhomogeneous broadening contribution.

The increase of the rates $T_1^{-1}$ and $T_2^{-1}$ in Fig. 3 with carrier concentration constitutes clear signature for electron-impurity scattering. $T_1^{-1}$ depends only weakly on the doping concentration, indicating that LO phonon scattering dominates over impurity scattering. Assuming a linear dependence of the impurity scattering rate $\tau_{\text{imp}}$ on the doping concentration in our well-doped structures, linear extrapolation gives rise to $T_1^{-1} = 2.13$ THz at $N_D = 0$, with a slope of $\tau_{\text{imp}} = 0.45 N_D$ cm$^2$/s. In particular, this means that about 22% of the intersubband relaxation rate is caused by impurity scattering for the highest doped sample. For the modulation-doped sample, however, $T_1^{-1}$ is as low as 1.56 THz, significantly below the extrapolated value. Taking account of the otherwise identical MBE growth parameters, the latter observation is unexpected and not understood at present.

The dephasing rate $T_2^{-1}$ found to depend more strongly on the doping concentration than $T_1^{-1}$. Here linear extrapolation for the well-doped case yields $T_2^{-1} = 5.6$ THz at $N_D = 0$. The slope (6±1N_D cm$^2$/s) is mainly associated with (intersubband) ionized impurity scattering, with some additional contribution from electron-electron scattering [because density dependent $T_2^{-1}$ is known to exist also in modulation-doped QWs (Ref. 10)]. Again, the dephasing rate for the modulation-doped structure is somewhat lower than expected from linear extrapolation.

| TABLE I. Doping concentrations (nominal and as determined by SIMS), measured transition energy $E_2 - E_1$, and peak wavelength $\lambda_{\text{peak}}$. Lorentzian linewidth $\Gamma_2$, and relaxation times $T_1$ and $T_2$ of the investigated samples. |
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| Sample | Nominal/SIMS doping ($10^{11}$ cm$^{-2}$) | $E_2 - E_1$ (meV) | $\Gamma_2$ (meV) | $T_1$ (fs) | $T_2$ (fs) |
| 1 | 4/4.4 (well doped) | 156.6 | 9.8 | 420 | 130 |
| 2 | 8/6.6 (well doped) | 163.2 | 11.0 | 410 | 100 |
| 3 | 16/13.4 (well doped) | 162.9 | 14.0 | 360 | 75 |
| 4 | 2/1.9 (mod doped) | 150.0 | 4.4 | 640 | 260 |
induced broadening, since this process should only affect the coherence time and be of negligible influence for intersubband scattering.

To check the temperature dependence, we have also performed intensity autocorrelation measurements on sample 1 at higher temperatures up to 150 K (not shown) and found only a slight decrease of $T_1$ from 420 to 390 fs. This is consistent with the expected behavior of emission rates associated with the Fröhlich interaction.$^{10}$

In conclusion, femtosecond dynamics of intersubband transitions in In$_{0.10}$Ga$_{0.90}$As/Al$_{0.31}$Ga$_{0.69}$As QWs has been studied by interferometric two-photon photocurrent autocorrelation measurements. The approach has been used to investigate population and phase relaxation times, in particular, their dependence on impurity concentration and impurity location, and to discriminate between different scattering processes. Knowing these dependencies will be crucial for further optimization of intersubband detectors, emitters, and modulators.

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