Time-Resolved Measurement of Intersubband Population Dynamics

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We apply an interband pump/intersubband probe technique to monitor the temporal evolution of the electron population in the first and second subband of an undoped GaAs/AlGaAs asymmetric double quantum well after interband optical excitation. The spacing between the two subbands is smaller than the longitudinal optical phonon energy. The time dependence of the intersubband absorption can be explained by a simple rate equation model. We extract an intersubband lifetime of T_{21} = 14 ps at an excitation density of $n_S = 1 \times 10^{10}$ cm^{-2}.

Introduction

Semiconductor nanostructures are fascinating quantum systems, which allow engineering of wavefunctions and transition energies. The development of quantum cascade lasers [1] has shown that applying quantum mechanics can lead to new optoelectronic devices. Several other novel devices have been proposed employing intersubband transitions in semiconductor nanostructures [2]. For all these proposals, the population dynamics and the dephasing times are the most crucial parameters. Ultrashort optical pulses with a large spectral width make it possible to study dephasing and relaxation times of carriers in the subbands of quantized structures. The optical pulses can be used to generate ultrashort broadband mid-infrared (MIR) pulses making it possible to measure the complete time-resolved absorption spectrum of a sample with a single pulse, offering simultaneously high resolution in both the time and frequency domains. In our experiment, an interband pump pulse injects electrons into the first and second subband of an undoped asymmetric double quantum well (ADQW) with a level spacing smaller than the LO phonon energy. The time evolution of the electron population in these two subbands is monitored by probing the MIR intersubband transitions to a third (empty) subband. The direct measurement of the subband populations allows us to determine whether there is intersubband THz gain in optically pumped structures.

Experimental

The ADQW sample was grown by molecular beam epitaxy on a semi-insulating (SI)-GaAs substrate. It consists of 40 periods of undoped GaAs wells with widths of 75 Å and 65 Å, separated by a 25 Å $Al_{0.30}Ga_{0.70}As$ barrier. The separation between each double well is 200 Å. A 100 Å AlAs sacrifice layer was grown between the substrate and the ADQW epilayer film. The sample was cleaved into a 1×1 mm^2 piece and etched for about 6 h in 10 % HF. After the QW film had been lifted off the substrate it was bonded via van der Waals forces to the base of a ZnTe prism. The prism forms a waveguide for the mid-infrared probe pulse and enables a considerable electric field component along the growth axis to achieve a strong coupling to the intersubband transition dipoles. Moreover, the ZnTe prism is (in contrast to GaAs) transparent for the probe as well as for the pump light, which allows collinear pumping to achieve a high time-resolution. The difference between the refraction indices of ZnTe in the near- and mid-infrared regions, however, leads to a walk-off between the pump and the probe

pulses that typically amounts to 150 fs. This is the time resolution that can be achieved in this geometry.

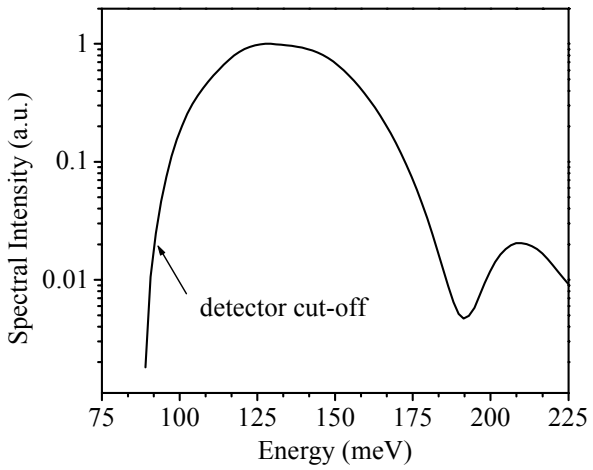

Fig. 1: Intensity spectrum of the MIR probe pulse generated by phase-matched difference frequency mixing in GaSe.

In our time-resolved photoinduced absoption (PIA) setup we use a mode-locked Ti:sapphire laser that delivers 12 fs pulses at a repetition rate of 75 MHz with an average output power of 800 mW. The laser pulses are centered at a wavelength of 780 nm, and the bandwidth is 110 nm (full width at half maximum). The laser beam is split into three parts: *pump*, *probe* and *analysis* beam. The *pump* beam is sent through a variable delay stage and is then used to excite the sample. The desired pump-pulse spectrum is selected through a grating pulse shaper. The sample is mounted on the cold finger of a He continuous flow cryostat and cooled to approximately 5 K. The *probe* is focused on a 30 µm GaSe crystal to generate the linear polarized MIR probe pulse through phase-matched difference mixing [3]. To satisfy the phase-matching condition the polarization of the laser pulse is rotated with a half-wave plate by about 45° out of the horizontal. By changing the orientation of the GaSe crystal it is possible to generate p- or s-polarized light. The MIR radiation transmitted through the sample is then focused on a liquid nitrogen cooled HgCdTe detector. This part of the setup corresponds to a conventional pump-probe experiment in which the total energy of the probe pulse is measured as a function of time delay after excitation. However, the determination of the electric field of the probe pulse requires heterodyne detection. This can be achieved by generating a quasi-single-cycle MIR pulse through optical rectification of the 12 fs *analysis* pulse in a 100 µm ⟨110⟩ SI-GaAs sample. In order to obtain time-resolution, the probe and analysis pulses are mixed at a Si beamsplitter and their superposition is detected as a function of time delay with the time-integrating HgCdTe detector. The signal is collected with a lock-in amplifier phaselocked to an optical chopper which modulates either the pump beam (PIA scan) or the analysis beam (reference scan). Figure 1 shows the intensity spectrum of the MIR probe pulse obtained through Fourier transform of the cross-correlation signal.

Results and Discussion

Figure 2 (a) shows intersubband absorption spectra taken at different time-delays after the excitation by an interband pump pulse. The spectra clearly exhibit two absorption peaks: one around 112 meV, the other one 14 meV above this value. The low-energy peak is due to the (2-3) intersubband absorption, while the second peak is attributed to

the (1-3) absorption. The amplitude of the first peak decreases with time-delay after excitation due to intersubband relaxation, while the amplitude of the second peak first rises slightly, and subsequently decreases due to carrier recombination. Since the area under the peak (i-3) (i = 1, 2) is directly proportional to the subband population $n_i(t)$, we are able to determine the population dynamics in the quantum well on the basis of the time-resolved absorption spectra [4]. Fig. 2 (b) shows the electron population of the first and second subband as a function of time delay after optical excitation (symbols). About 40% of the photo-excited carriers (1 x 10^{10} cm^{-2}) are injected into the second subband, while the remaining 60% are injected into the first subband at higher k-value. The population in the second subband (squares) shows an exponential decay. The carriers relaxing down from the second subband add to the population in the ground level. Subsequently, the population in the ground level drops due to carrier recombination. The lines in the inset of Fig. 2 (b) are the results obtained from a simple rate equation model. By fitting the calculation to the experimental data we deduce an intersubband relaxation time of T_{21} = 14 ps.

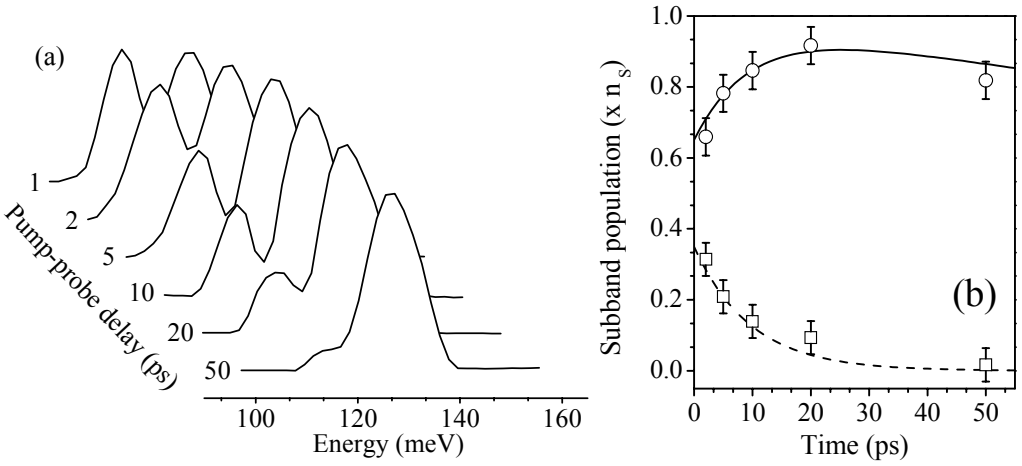

Fig. 2: (a) Time-resolved intersubband absorption spectra for the ADQW sample.
(b) Population in the first (solid line) and second (dashed line) subband.

We will now discuss possible intersubband scattering mechanisms and compare numerical estimates of scattering rates with the experimental results. Since the subband spacing in our sample is smaller than the LO phonon energy the electrons in the second subband do not possess sufficient energy to emit LO phonons and intersubband relaxation can only be due to acoustic phonon emission and electron-electron (e-e) scattering. We calculated the acoustic phonon scattering time to be some hundred ps for our structure. This time is much too long to explain our experimental findings. Recent calculations, however, show that the intersubband e-e scattering rates can be very high, almost approaching in some circumstances the intersubband scattering rate due to LO phonon emission [5]. We calculated the e-e scattering rates in the Born-approximation using static single subband screening within the random phase approximation. The most prominent intersubband e-e scattering processes are labeled 2211 and 2221, where *ijfg* describes an interaction, where an electron in state *i* scatters to *f* under collision with a second electron, which scatters from *j* to *g*. When working out the total population transfer rate between the first and second subband, both scattering processes and the number of electrons that are transferred by each process were taken into account. We calculated an intersubband e-e scattering time of $T_{21} = (2\ W_{2211} + W_{2221})^{-1}$ = 18 ps which is in good agreement with the experimental result. Some authors have also observed a strong reduction of the intersubband relaxation time due to

fast relaxation of electrons in the high-energy tail of the hot carrier distribution in the second subband by emission of LO phonons [6]. Due to the small subband spacing of our sample and the low excitation density, however, the injected electron population in the second subband is cold enough that LO phonon emission is suppressed. Finally, we should also mention our study of the excitation density dependence. We have measured photoinduced intersubband absorption spectra when varying the excitation density from 1 x 10^{10} to more than 1 x 10^{12} cm^{-2}. With increasing densities we observe a significant shortening of the relaxation times. Whereas, in the high density regime we observe a steep increase of the intersubband relaxation times (due to Pauli blocking of the final states). A similar excitation density dependence has been observed by Hartig et al. in a time-resolved photoluminescence experiment [7].

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