Femtosecond pump-probe spectroscopy of intersubband relaxation dynamics in narrow InGaAs/AlAsSb quantum well structures

C. V.-B. Tribuzy, a,1 S. Ohser, S. Winnerl, J. Grenzer, H. Schneider, and M. Helm
Institute of Ion Beam Physics and Materials Research, Forschungszentrum Rossendorf, P.O. Box 510119, 01314 Dresden, Germany

J. Neuhaus and T. Dekorsy
Department of Physics, University of Konstanz, 78457 Konstanz, Germany
and Center for Applied Photonics, University of Konstanz, 78457 Konstanz, Germany

K. Biermann and H. KüNZel
Fraunhofer Institute for Telecommunications, Heinrich Hertz Institut, 10587 Berlin, Germany

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Intersubband relaxation dynamics in InGaAs/AlAsSb multiquantum wells (QWs) is studied by single-color femtosecond pump-probe measurements. At early delay times, all samples show an exponential decay of the transient transmission occurring with time constants of the order of a picosecond. The relaxation dynamics at later delay times strongly depend on both QW thickness and doping location. A non-single-exponential decay behavior indicates extra competing relaxation channels, as further confirmed by solving three-level rate equations. It is shown that slowly decaying components are due to electron transfer to states related to indirect valleys in the wells or in the barriers. © 2006 American Institute of Physics. [DOI: 10.1063/1.2360242]
samples B and C at 0.51 eV. Due to the higher doping, sample A exhibits a larger integrated absorption than samples B and C.

For the pump-probe measurements, femtosecond optical pulses with about 240 fs duration were generated at 78 MHz repetition rate by an optical parametric oscillator tunable from 1.3 to 3.2 μm. A small angle between the pump and probe beams, which were polarized parallel to the growth direction, was used in order to separate both beams. For the measurements a scanning delay generator (shaker) between pump and probe beams was operated at a frequency of 48 Hz and the signal was accumulated with a fast analog/digital converter (fast-scanning technique). All measurements were performed at room temperature. The pump-pulse energy was about 130 pJ at a wavelength of 2.4 μm and 70 pJ at 3.1 μm, exciting only about 1% of the electrons into the excited state. As discernible in Figs. 2(a)–2(c) below, the high pulse repetition rate together with the fast-scanning technique provides an unprecedented signal quality. Transmission changes of 1% are measured with a signal-to-noise ratio of up to 1000, making absorption changes of 10^{-5} detectable, several orders of magnitude better than reported in previous intersubband relaxation measurements.

Figure 2(a) shows the relative probe transmission change ΔT/T₀ for sample A. At very early delay times we observe a coherent artifact arising from self-diffraction of the pump into the direction of the probe beam, which will not be discussed here further. At later time delays, the decay of the signal can be accurately fitted by a single exponential, yielding a decay time constant of τ = 1.5 ps.

The band structure of our samples was calculated by a self-consistent solution of the Schrödinger and Poisson equations, including nonparabolicity as described in Ref. 19 and using an InGaAs band edge mass of 0.0427m₀ at the Γ point. m₀ is the free electron mass. Other parameters are taken from Ref. 20. Figure 2(d) shows the conduction band edge profile of sample A at the Γ and X minima (for simplicity, the L valley is not shown), as well as the square of the modulus of the lowest two subband wave functions at both minima. Even though the second Γ subband coincides approximately with the X levels for the chosen band parameters, the pump-probe data appear to be unaffected by intervalley scattering.

Studying the relaxation time of sample B, its pump-probe curve is best fitted by a single-exponential plus a constant offset, as shown in Fig. 2(b). The offset is necessary to describe the slow decay (nanosecond regime) observed at later delay times. For sample B a decay time constant of τ = 0.8 ps was found. As plotted in Fig. 2(e), the excited Γ subband is raised significantly above the X levels due to the reduced well width. In addition, the excited carriers can now easily escape into the barriers, where metastable states at the X minimum exist due to the barrier doping. The extremely slow relaxation at large delays indicates the relevance of the
latter process. In fact, these barrier-confined metastable states exhibit negligible tunneling probabilities due to the large effective mass at the indirect minimum and a high barrier (about 0.4 eV) for thermionic emission back into the QW.

Comparing these decay time constants, sample B should exhibit a longer intersubband relaxation time than sample A due to its larger intersubband energy and the $1/q^2$ dependence of the optical phonon scattering, where $q$ is the momentum transfer. The observed opposite behavior is therefore another signature for the additional relaxation channels present in this sample.

In order to eliminate long-lived states in the barrier, we now turn to sample C where the Si impurities are located inside the QWs and space charge effects are negligible [see Fig. 2(f)]. The resulting pump-probe data are shown in Fig. 2(c). Indeed the plateau-like component at later decay times is not present. Nevertheless a slow component is also observed. In fact, the pump-probe curve of this sample is best fitted by a biexponential decay without offset. The two characteristic decay time constants are 1.2 and 6.2 ps. According to Fig. 2(f), levels related to indirect valleys inside the InGaAs QW are the only possible origin of the long time constant.

We have also performed a rate equation analysis involving three levels [shown in the inset of Fig. 2(e)] to describe the two decay time constants. Assuming one effective level $(X)$ for the $X$ and $L$ states, the longer decay time $\tau_2$ then characterizes the return time from the $X$ or $L$ levels to level 1, the fundamental $\Gamma$ subband. This return time is expected to be longer than the scattering time $\tau_X$ from level 2 into the side valleys due to the higher effective masses of the latter.\(^1\) The initial decay time $\tau_1$ is thus attributed to the combined scattering between the two $\Gamma$ levels of the QW ($\tau_2$) and the one related to $\Gamma-X$ or $\Gamma-L$ transfer ($\tau_X$), leading to the relation $1/\tau_1 \approx 1/\tau_2 + 1/\tau_X$. Even though $\tau_2$ and $\tau_X$ cannot be determined independently, both time constants have to exceed the observed value of $\tau_1$ (1.2 ps). In fact, this conclusion is further supported by the observation that this value of the “fast” decay time is shorter than for the configuration of Fig. 2(d) where intervalley scattering has only negligible influence and longer than for Fig. 2(e) where emission into the barrier provides an additional channel. Knowing that typical $\Gamma-X$ scattering times for bulk III-V materials are of the order of 100 fs,\(^2\) our observations indicate that intervalley scattering times in QWs should be significantly longer than for bulk material, as also suggested in Refs. 23 and 24, however, depending very strongly on the exact wave functions involved. Our observations are sustained by the fact that pronounced intersubband luminescence has been observed from states located somewhat above the side valleys,\(^2\) which implies a reasonably long intervalley scattering time.

In conclusion, pump-probe measurements on InGaAs/AlAsSb QWs at low excitation densities were carried out in single-pass geometry with very high signal-to-noise ratios. At early decay times, all samples show an exponential decay of the transient transmission occurring with time constants of 0.8–1.5 ps. The relaxation dynamics at later time delays strongly depends on both QW thickness and doping location. For barrier-doped structures, very long time constants in the nanosecond regime can emerge if carriers escape into the barriers. In the case of well doping, intervalley scattering of high-energy electrons located above the indirect minima of the well material was found to occur with much longer time constants, in the picosecond regime, than for bulk material. In particular, the latter observation suggests that intersubband lasing involving states above indirect minima of the well material should be possible. This work additionally provides important details about the relaxation dynamics of short wavelength intersubband transitions relevant for all-optical intersubband modulators designed for telecommunication wavelengths and saturable absorbers for IR to mid-IR solid state or fiber lasers.

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