Direct monitoring of the excited state population in biased SiGe valence band quantum wells by femtosecond resolved photocurrent experiments

P. Rauter,[a] T. Fromherz, and G. Bauer
Institute of Semiconductor and Solid State Physics, University of Linz, Upper Austria 4040, Austria

N. Q. Vinh
FOM Institute for Plasma Physics Rijnhuizen, 3430 BE Nieuwegein, Netherlands

B. N. Murdin
University of Surrey, Guildford, Surrey, GU2 7XH United Kingdom

J. P. Phillips and C. R. Pidgeon
Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom

L. Diehl, G. Dehlinger, and D. Grützmacher
Paul Scherrer Institut, 5253 Villingen, Switzerland

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The authors report a *direct* measurement of the optical phonon intersubband hole relaxation time in a SiGe heterostructure and a quantitative determination of hole relaxation under electrically active conditions. The results were obtained by femtosecond resolved pump-pump photocurrent experiments using a free electron laser (wavelength 7.9 μm). Additionally, the intensity dependence of the nonlinear photocurrent response was measured. Both types of experiments were simulated using a density matrix description. With one parameter set, a consistent modeling was achieved confirming the significance of the extracted heavy hole relaxation times. For an intersublevel spacing of 160 meV, a value of 550 fs was obtained. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2397004]

Driven by the strong need for cheap and integrable Si-based optoelectronic devices for a wide range of applications, considerable endeavors have been made to develop structures for light emission, modulation, and detection in this material system. While recent breakthroughs like the demonstration of a high-speed optical modulator in Si (Ref. 1) bring the concept of transition from electrical to optical interconnects closer to realization, the base for any silicon photonics, namely, a group IV laser source, still has to be developed. Up to now, the only lasing devices demonstrated in Si are a Raman laser[2] and Si-based impurity lasers,[3] which essentially lack the advantages associated with the silicon system by requiring an external pump laser source. For silicon as an indirect semiconductor the concept of infrared emitters based on quantum cascade (QC) heterostructures, which is very successfully applied to III-V material systems, constitutes a promising approach towards a SiGe infrared laser. But while infrared electroluminescence (EL) of various wavelengths has been demonstrated for *p*-type SiGe quantum cascade structures,[4–6] lasing has yet to be achieved. The buildup of population inversion in order to achieve lasing fundamentally depends on the relaxation lifetime of the excited energy level of the lasing transition. Therefore the measurement and optimization of the intersubband relaxation time constitutes a key issue for the realization of a Si cascade laser source. The intersubband hole relaxation time for transitions *below* the optical phonon energy (Si–Si: 58 meV, Ge–Ge: 36 meV) is significantly larger than 10 ps and therefore is experimentally well accessible.[7] The lifetimes for energies *above* the optical phonon energies are smaller than 1 ps. Thus so far experiments suffered from a lack of time resolution[8] or from hole heating requiring subtraction of heating contributions in order to gain a value for the relaxation time.[9]

In this work we report the first direct measurement of the excited heavy hole (HH) lifetime in the femtosecond regime for quantum well (QW) transition energies above the optical phonon energy. Time resolved measurements were performed using the free electron laser (FEL) FELIX at FOM Rijnhuizen. FELIX provides micropulses with full widths at half maximum down to 280 fs and peak powers of 100 MW. The micropulse repetition rate is 25 MHz, five macropulses of 7 μs duration are provided per second. The FEL power can be reduced to 33 dB by attenuators.

The sample was grown pseudomorphically on a Si [100] substrate. The active region contains ten periods of nominally the same five coupled SiGe QWs, for that QC EL was observed recently.[4] The periods are separated by 500 Å undoped Si barriers in order to suppress dark current. A detailed description of the sample investigated in this work is given in Ref. 10. In Fig. 1(d), the band structure of this sample as calculated according to Ref. 11 is shown for the HH, light hole, and split-off band edges. The absolute squares of the wave functions relevant for this work are shown at their respective energy values. The HH1-HH2 transition energy of the deepest well 1 (39 Å, 42% Ge) is calculated to be 160 meV. Experimentally, this transition energy was determined by voltage modulated waveguide transmission. The occupation of the HH1 state was modulated via an externally applied ac voltage (±2 V, 50 kHz) and the resulting change

[a]Electronic mail: patrick.rauter@jku.at

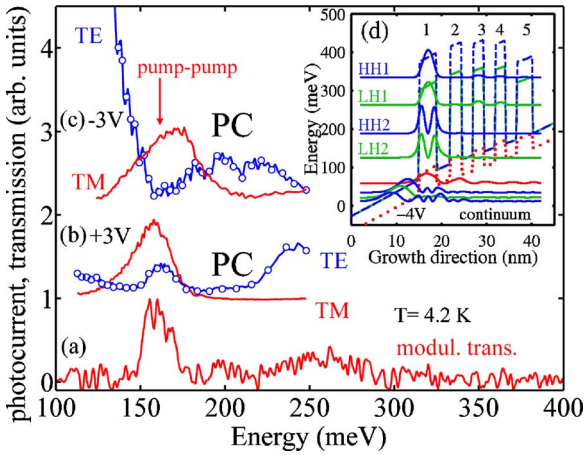

FIG. 1. (Color online) Absorption, two-photon PC, and simulation results. Inset (d) shows the calculated heavy, light, and split-off (broken, solid, and dotted line) hole band edges at a bias of −4 V and a plot of the relevant absolute squared wave functions at their eigenenergy values. The calculated HH2-HH1 energy difference (160 meV) is consistent with the peak positions measured in voltage modulated TM transmission experiments (a) and in two-photon PC spectra in TE and TM polarizations (line with and without symbols) obtained by varying the FEL wavelength [(b) and (c) for ±3 V bias]. The graphs were offset vertically for clarity. The arrow labeled pump-pump indicates the energy at which the time resolved measurements were performed.

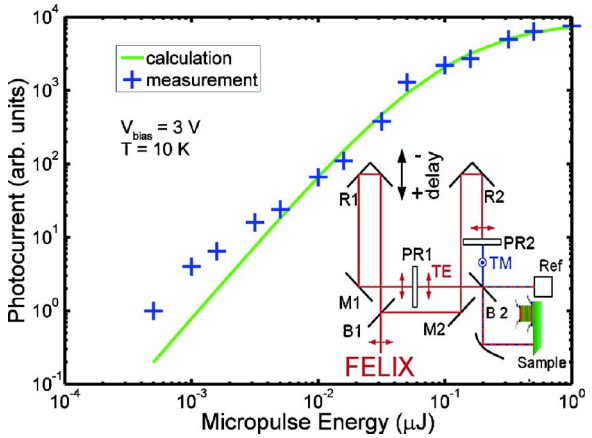

FIG. 2. (Color online) Double logarithmic plot of the PC signal vs the macropulse power of FELIX. The experimental data (crosses) were measured at a bias of 3 V and a temperature of 10 K in TM polarization. The simulated dependence of the PC is shown as solid line. The inset shows the pump-pump setup. A detailed description is given in the text.

of the spectrally resolved waveguide transmission was measured via lock-in technique for transversal magnetic (TM) radiation. Figure 1(a) shows that a clear peak at the calculated HH1-HH2 transition energy is observed. The final HH2 state of this transition is strongly confined. Thus the HH1-HH2 transition of the deepest QW 1 was not observed in our previous photocurrent (PC) experiments,[10] in which low intensity radiation from a globar was used to excite the sample. However, since in well 1 the HH1, HH2, and the onset of the continuum states are nearly equally spaced in energy, PC can be generated by two-photon processes. Figure 1 shows the results of PC experiments using the intense laser radiation of FELIX. For two different bias voltages, Figs. 1(b) and 1(c) show a clear two-photon PC signal resonantly enhanced at the HH1-HH2 transition energy for TM polarization.

In order to verify that a nonlinear absorption process is responsible for the PC signal at the HH1-HH2 energy, the dependence of the PC peak maximum on the FEL micropulse energy was measured for TM polarized radiation. For pulse energies smaller than 3×10^{-2} μJ the results presented in Fig. 2 show a clear superlinear dependence of the peak maximum on the pulse energy typical for multiphoton processes. For pulse energies in excess of 5×10^{-2} μJ, a saturation of the PC signal is observed.

In order to understand the power dependence of the PC signal over the whole range of micropulse energies as well as the dynamics of the state occupancies, the response of the system to a laser pulse resonant with the HH1-HH2 transition was simulated using a density matrix (DM) approach.[12,13] The result of the DM calculations is shown by the solid line in Fig. 2. The details of the DM calculations are beyond the scope of this letter and will be published in a forthcoming publication.[14] The simulation is in very good agreement with the measurement over two orders of magnitude of the exciting power. The calculations show that for low excitation intensities HH2 is only weakly populated by the pulse, most carriers remain in HH1. Therefore the continuum occupation depends quadratically on the intensity. As the peak intensity gets higher, the HH1-HH2 transition reaches bleaching (i.e., the HH1 and HH2 populations become equal) within the pulse time, and therefore the continuum occupation and the PC become linearly dependent on the excitation intensity. For even higher intensities, the occupations of HH1, HH2, and continuum become comparable, thus the transition to the continuum becomes bleached and the PC saturates.

In order to determine the HH2-HH1 relaxation time, which has been reported to be mainly determined by optical phonon deformation potential scattering,[9,15] PC pump-pump experiments were performed. The pump-pump setup is shown in the inset of Fig. 2. The FEL beam with a wavelength of 7.9 μm enters the optical setup in TE polarization and is split by a beam splitter (B1). The polarization of one of the beams is turned by 90° into TM polarization (PR2). The beam remaining in TE polarization is reflected by a movable mirror (R1) allowing to adjust the delay between TE and TM pulses on a femtosecond scale. Before being coupled into the sample waveguide, the beams are made collinear again by the use of a second beam splitter (B2). The integral PC through the variably biased sample originating from the delayed TE and TM micropulses is measured as a function of the delay. The advantage of performing PC measurements in comparison with pump-probe experiments lies in its much higher sensitivity. In addition, major setup problems arising for pump-probe transmission experiments in waveguide configuration, like distinguishing between the parallel pump and probe beams, are avoided. The cross polarization of the FEL beams breaks the symmetry of the experiment with respect to the sign of the delay and allows an unambiguous identification of decay processes even if the decay time constant is close to the time resolution of the setup. Furthermore it prevents interference of the two beams.

The results of the pump-pump measurements after subtracting a background that corresponds to the PC signal for large delays are presented in Fig. 3. The curves show a pronounced asymmetry with respect to the sign of the delay, which is due to the strong polarization dependence of the transition involved. For negative delays in Fig. 3, the TM polarized pulse is the first to interact with the sample. In this

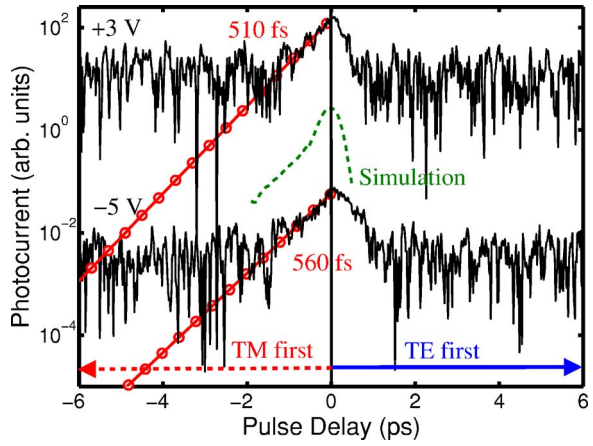

FIG. 3. (Color online) Logarithmic plot of the PC through the sample as a function of the delay between the TM and TE pump pulses for different bias voltages. A constant background equivalent to the current integral over the two noninteracting pulses has been subtracted and the traces have been offset vertically for clarity. The FEL macropulse powers were 160 μJ (TM) and 130 μJ (TE) attenuated by 5 dB for a bias of −5 V and 120 μJ (TM) and 160 μJ (TE) attenuated by 20 dB for a bias of 3 V. Note the pronounced asymmetry with respect to the sign of the delay. An exponential fit results in a HH2-HH1 relaxation time of about 550 fs independent of the bias (symbols). Also shown is the result of pump-pump simulations (broken line) using the same parameters as in the simulation of the power PC dependence on the micropulse energy shown in Fig. 2.

polarization, the HH1-HH2 transition is allowed, and thus a nonequilibrium population of the HH2 state will be excited. This nonequilibrium HH2 occupation decays exponentially with the time constant τ_{10}. From the HH2 state, holes can be excited to the continuum also in TE polarization. Therefore, if the TE pulse hits the structure before the HH2 carriers have relaxed, the integral PC increases proportionally to the residual holes in the HH2 state. Thus the measured decay of the integral PC as a function of the pulse delay directly monitors the HH2 lifetime. Since the HH1-HH2 selection rules prohibit a HH2 population generated by the TE pulse, the HH2 relaxation cannot be observed if the TE pulse arrives first (positive delays in Fig. 3) and an asymmetry with respect to the pulse order as observed in the experimental results. The fast decaying increase of the PC signal between 0 and 1.5 ps in Fig. 3 results from the overlap of the TE and TM pulses, and thus indicates the time resolution of the experimental setup. An exponential fit reveals a HH2-HH1 relaxation time of 550 fs, which is significantly longer than the time resolution of the measurement system. Figure 3 also shows the simulated results for pump-pump experiments.[14]

As shown by the broken line in Fig. 3, our simulation accurately describes the experiment. We want to point out that the same parameters as used for simulating the dependence of the PC peak maximum on the micropulse energy were used in the simulation of the pump-pump results. The experiment was performed for several bias voltages, however, no influence of the bias voltage on the HH2-HH1 relaxation time could be observed. From our experiments, a significantly longer relaxation time (550 fs) than reported in Ref. 9 (250 fs) results, where a contribution of hole heating had to be deconvoluted in order to gain this value. In our experiment, such heating effects would only enter via a time dependent development of the distribution function for the *excited* HH2 population into a Fermi-Dirac distribution described by an elevated hole temperature and a HH2 quasi-Fermi level. Calculations indicate a decrease of the HH2-continuum absorption at the pump-pump wavelength of 7.9 μm as a consequence of heating. Thus, a deviation from exponential decay and an *underestimation* of the HH2 relaxation time would result due to heating. However, we want to emphasize that due to the low excitation intensity sufficient for our experiments, no deviations from an exponential HH2 decay could be observed. Also in previous work, under the higher intensities required for pump-probe absorption determinations of lifetime, only the heating of the HH1 *ground* state distribution was subtracted in the analysis and the influence of a hot HH2 excited state distribution was neglected.[9]

The excellent agreement of measured and simulated results for both types of experiments with one set of parameters strongly supports the interpretation of our data. The extracted HH2-HH1 subband relaxation time τ_{10} is thus highly reliable, for it is consistent with the results of both types of experiments. To our knowledge this work presents the first *direct* measurement of the intersubband hole relaxation time for transitions above the optical phonon energy. Additionally, femtosecond time resolution has been achieved here for the first time in an active, electrically biased SiGe structure. Thus our results constitute a key step in the design and dynamic simulation of SiGe QC laser structures.

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