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Long intersubband relaxation times in *n*-type germanium quantum wells

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We measured the non-radiative intersubband relaxation time in *n*-type modulation-doped Ge/SiGe multi-quantum wells of different thickness by means of degenerate pump-probe experiments. The photon energy was tuned to be resonant with the lowest conduction band intersubband transition energy (14–29 meV), as measured by terahertz absorption spectroscopy and in agreement with band structure calculations. Temperature-independent lifetimes in excess of 30 ps were observed.

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Strained Ge/SiGe heterostructures have been recently identified as a possible solution to realize silicon-based emitters and amplifiers of radiation in the terahertz range^{1–3} based on intersubband transitions (ISBT). The use of nonpolar SiGe heterostructures instead of III-V compounds for the realization of quantum cascade lasers (QCLs) may help filling the wavelength gap between existing terahertz and mid-infrared QCLs, created by polar optical phonons (reststrahlen band). We have recently investigated *n*-doped strained Ge/Si_{1–x}Ge_x multi-quantum well (MQW) structures with Ge-rich ($x > 0.75$) barriers, demonstrating clear ISBT signatures in the 20–50 meV range, in good agreement with band structure calculations.^{4,5} Electron-doped Ge-rich heterostructures have been identified to be more promising than *n*-type Si rich system because of the smaller confinement effective mass of electron at L point.^{1,2} Furthermore, they permit to circumvent the difficulties of QCL designs based on *p*-type doped SiGe wells for which both the strong nonparabolicities of the different valence bands⁶ and the carrier scattering among them limit the non-radiative intersubband relaxation times τ . Indeed, previous experimental determination of τ in SiGe/Si heterostructures by time-resolved pump-probe transmission experiments was restricted to *p*-type systems,^{7–10} and very short $\tau < 1$ ps were reported^{8,9} for ISBT energies above the longitudinal optical phonon energies $\hbar\omega_{\text{opt}} = 37(63)$ meV for Ge(Si). At ISBT energies lower than $\hbar\omega_{\text{opt}}$, the main scattering channel was instead due to alloy disorder in the SiGe wells, with values of τ around 10 ps.^{7,10} Scattering by alloy disorder and rough interfaces should be strongly reduced in *n*-type Ge QWs with thickness $d_{\text{QW}} > 10$ nm (Ref. 11): long lifetimes, desirable for the QCL design, are then expected for such structures. We have thus grown *n*-type modulation-doped strained Ge/Si_{0.2}Ge_{0.8} MQW samples, and we have determined the non-radiative τ using narrow-band picosecond pulses provided by the Dresden free-electron-laser (FEL) FELBE. The laser photon

energy was tuned at the ISBT energy to measure the transient bleaching signal by means of far-infrared degenerate pump-probe measurements.

The SiGe heterostructures were grown by means of ultra-high vacuum chemical vapour deposition from high purity silane and germane on Si(001) substrates. The MQW structures were deposited at 500 °C on a partially relaxed reverse graded virtual substrate grown on silicon,^{12,13} having final Ge contents $x = 0.8$. The MQWs were made of 20 repetitions of Ge/Si_{0.2}Ge_{0.8} with $d_{\text{QW}} = 13$ nm (sample S1776) or $d_{\text{QW}} = 24$ nm (S1745 and S1750). The 30 nm-thick Si_{0.2}Ge_{0.8} barriers were doped by phosphine co-deposition resulting in a dopant concentration of $N_d \sim 1 \cdot 10^{18} \text{ cm}^{-3}$. The barriers were either doped uniformly or alternatively 10-nm thick Si_{0.2}Ge_{0.8} spacers were left undoped on each side of the QW (see Table I).

Electronic states calculations were performed by means of an effective-mass self-consistent approach,⁴ where states originating both from the L and Δ_2 band edges are included in the model, since they have comparable energies.¹⁴ Γ and Δ_4 states are much higher in energy and can be disregarded in this context. As an example we show in Fig. 1 the band edge profiles and the square modulus of the near-gap states for the conduction band of sample S1776 calculated at $T = 10$ K. The QW ground state $L0$ and first excited state $L1$ are clearly visible. In the doped portion of the barrier, the donor levels lie 24.7 meV below the Δ_2 bands. We find that for the chosen temperature the donors are partially ionized, so that only a fraction of the doping charge is transferred into the well.

Low temperature (T) absorption spectra were measured by FT-IR in order to identify the ISBT energy. The radiation was coupled to the samples by adopting a waveguide geometry with lateral facets at 70° with respect to the growth plane. The top (growth) side was coated with a 5 nm-thick Ti layer followed by an 80 nm-thick Au metallization in order to ensure a good overlap of the optical mode with the active MQWs region.⁴ The waveguide length and thickness were designed to allow only one light reflection inside the waveguide, as required for a precise determination of τ .¹⁵ The

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TABLE I. Parameters and results for the samples discussed in this work. The ISBT energy is determined from both FT-IR experiments (exp) and band calculations (theo). τ is the non-radiative relaxation time as determined from pump-probe experiments. The TM-mode transmittance $T_{\text{TM}(v)}$ at the used FEL photon energy is also reported.

Sample	d_{QW} (nm)	Spacer	ISBT theo (meV)	T (K)	ISBT exp (meV)	$h\nu_{\text{FEL}}$ (meV)	$T_{\text{TM}(v)}$	τ (ps)
S1745	24	no	15	4	16.2	14.1	0.71	27
				30	17.9	14.1	0.71	34
				90	20	18.2	0.66	30
S1750	24	yes	13	30	14.7	14.1	0.67	34
				90	18.0	18.2	0.64	34
S1776	13	yes	32	4	28.4	28.8	0.73	33
				50	28.5	28.8	0.72	33
				130	28.5	28.8	0.71	33

dichroic transmittance shown in Fig. 2 is obtained as the ratio between the transmitted beam intensity with non-vanishing component of the polarization parallel to the growth direction (TM mode) and the transmitted intensity with orthogonal polarization (TE mode). One single absorption feature with Lorentzian lineshape and full width of about 3(5) meV was observed in samples with (without) undoped spacers at all T up to 130 K. As shown in Table I the measured ISBT energies agree well with the theoretical values obtained including the depolarization shift.⁴

The intersubband non-radiative lifetimes were measured in a degenerate pump-probe scheme¹⁵ with the FEL emitting a continuous train of pulses (duration from 4.8 ps for $h\nu_{\text{FEL}} = 28.8$ meV to 13 ps for 14.1 meV, with separation of 77 ns). The pump power was reduced by a set of attenuators to 40 mW (pulse energy ~ 3 nJ, power density at focus around 6 kW/cm^2). The transmission change Δ relative to the absolute TM-mode transmittance $T_{\text{TM}(v_{\text{FEL}})}$ obtained from FT-IR data (so that $|\Delta| < 1$) was determined as a function of pump-probe delay δt .

All curves presented in Fig. 3 show $\Delta > 0$ (bleaching) indicating a decreased population of the lower subband at $\delta t = 0$ and subsequent relaxation towards equilibrium.^{15,16} In Fig. 3(a) we plot the measured Δ for sample S1745. The exponential decrease of Δ vs. δt is extremely clear at all measured T and the exponential fitting of each curve in Fig. 3(a) provides the non-radiative relaxation time τ with relative

uncertainty below 5% (see Table I). In Fig. 3(b) we plot Δ for all three samples (T = 30 K for S1745 and S1750, T = 50 K for S1776); exponential decays with very similar relaxation times are obtained. We found values of the order of $\tau \sim 30$ ps independent of the value of T and d_{QW} and of the presence of a spacer layer. We stress that a T-independent value of τ is beneficial for high-T operation of QCLs (Refs. 1, 3, and 8) (note that τ^{-1} is an exponentially decreasing function of $1/T$ in III-V QCL heterostructures with single-well transition¹⁷).

We shall now consider all scattering processes separately. First of all we note that in all our samples the energy difference $E_{L1} - E_{L0}$ is smaller than the Ge-Ge optical phonon energy $\hbar\omega_{\text{opt}} = 37$ meV. Therefore, optical phonon scattering is strongly suppressed^{11,16,18} for S1745 and S1750, while it might play a role in S1776, where $E_{L1} - E_{L0}$ is only 8 meV lower than $\hbar\omega_{\text{opt}}$. Since we did not observe a shorter τ for S1776, the optical phonon scattering channel seems not to contribute to the observed lifetime. A second phonon-mediated scattering channel may be related to intervalley transitions¹¹ between L1 and the quasi-continuum of Δ_2 states, represented by red curves in Fig. 1. Note that for sample S1776 the level L1 is located in an energy range where Δ_2 levels are also present, while samples with a larger d_{QW} (S1745 and S1750, not shown in Fig. 1) have the first excited L level well below the Δ_2 continuum. If the value of τ would

FIG. 1. (Color online) Self-consistent band edge profiles (thick lines) and squared modulus of the wavefunctions (thin lines) calculated for sample S1776 at T = 10 K in the absence of optical excitation. Profiles and states at L-point (black lines, confined states L0, L1) and at Δ_2 -band edge (red lines) are displayed and marked. The donor level in the doped portion of the barrier is also represented (blue line). The green shaded area represents the energy region from the Fermi level to $2k_B T$ above it.

FIG. 2. (Color online) TM to TE waveguide mode transmittance ratio for two samples at T = 10 K (thin lines) and T = 90 K (thick lines). Lorentzian-shaped dips are the signature of the L0 $\rightarrow$ L1 intersubband transition. Spectra are vertically offset in steps of 0.4 for clarity. Data around 41 meV for T = 10 K are discarded, as the transmitted signal is zero because of absorption by donor impurity levels in the Si substrate.

FIG. 3. (Color online) Relative transmission change Δ (see text) measured with the free electron laser photon energy tuned close to the ISBT energy for each sample and temperature (see Table I). A fit to a single-exponential decay provides the intersubband transition lifetimes τ . (a) Temperature dependence for S1745. (b) Results for different samples at $T = 30$ K ($T = 50$ K for S1776). The maximum value of Δ is indicative of the fraction of confined electrons excited by the pump pulse.

be dominated by $L1-\Delta_2$ scattering, we should observe large variations in τ among samples with different d_{QW} . Since the observed values are approximately sample independent we conclude that the time scale is not set by the intervalley scattering. Indeed the Δ_2 states are localized in a different spatial region. Therefore the phonon-assisted intervalley $L1-\Delta_2$ scattering processes are strongly suppressed due to the small overlap factor^{1,18} which we found to be 2 orders of magnitude smaller than the corresponding quantity evaluated by using $L0$ as the final state. Concerning other inelastic interactions, we are left with acoustic-phonon intersubband scattering followed by hot-carrier thermalization,^{16,18} which is thought to play a key role in group-IV conduction-band materials at electron energies well below $\hbar\omega_{opt}$.^{1,2,11} However the predicted temperature dependence of the phonon-mediated relaxation time is not observed in our measurements, indicating that also the acoustic-phonon channel is not the dominant one.

Elastic scattering channels due to static potential energy perturbations such as interface roughness, ionized impurities and/or alloy disorder can also contribute to intersubband relaxation, in conjunction with fast intrasubband thermalization.^{3,11} The temperature independence of the measured τ would suggest that such elastic scattering processes dominate the intersubband relaxation. However, since we used a modulation-doped scheme, impurity scattering rates should be strongly dependent on the presence of undoped spacers. Moreover alloy disorder effects, which in pure-Ge wells are active only in the barrier region, and interface roughness scattering should both depend on d_{QW} .¹¹ The fact that these dependences are not measured in our samples (see Fig. 3(b)) suggests that the dominant intersubband scattering channel is related to static potential energy perturbations *inside* the Ge well, such as dislocations and background impurities. If this

is the case, even longer relaxation times could be achieved by optimizing further the sample growth process.

In conclusion we have measured the non-radiative intersubband relaxation times for electrons in Ge quantum wells displaying intersubband transition energy between 14 and 29 meV, i.e., within the frequency range of the reststrahlen absorption of typical III-V semiconductors used for existing quantum cascade lasers. We obtained relaxation times up to 33 ps at 130 K nearly independent of temperature and well width. Our results indicate that n-type Ge/SiGe heterostructures are very promising for the realization of Si-based emitters in a wide far-infrared range, eventually capable of high temperature operation.

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