Hole system heating by ultrafast interband energy transfer in optically excited Ge/SiGe quantum wells

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An efficient scattering process between the electron system in the L valley and the hole system in the Γ valley in Ge/SiGe quantum wells is identified. Its dependencies on excitation energy and carrier density are analyzed using spectrally and time-resolved pump-probe experiments. This carrier scattering causes an ultrafast heating of the hole system leading to an additional bleaching signature appearing a few tens of picoseconds after the excitation. Our findings are supported by microscopic calculations of the absorption spectra for various carrier densities and temperatures based on the semiconductor Bloch equations. Additionally, this scattering mechanism explains the enhanced free carrier absorption observed in previously reported pump-probe experiments.

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I. INTRODUCTION

In recent years, Ge has gained more and more relevance in the Si photonics community. The main reason for this increased interest is its well-established epitaxial growth on Si combined with a quasidirect band gap. Due to the latter, Ge exhibits a characteristic response following optical excitation similar to typical III-V materials, enabling the optical functionalization of Si. Recently observed phenomena in Ge/Si materials include transient optical gain, lasing, and the quantum-confined and dynamical Stark effect.[1–4] In order to develop integrated optical devices based on these phenomena, extensive studies of the carrier dynamics in this material system are indispensable.

The investigation of the optical properties of Ge has a long history starting in the beginning of the 1950s with the determination of its coefficients of extinction and refraction.[5–7] In 1974, first optical analyses of the carrier dynamics in Ge were performed by monitoring the hot electron system using picosecond laser pulses in a pump-probe setup.[8] Successive experiments addressed various cooling processes influencing the light-matter coupling of optically excited Ge, such as free carriers and interband transitions,[9,10] carrier diffusion,[11] intervalley scattering and thermalization of conduction-band electrons,[12–14] inter- and intra-valence-band scattering of holes,[15–17] and nonequilibrium phonon populations.[18,19] The carrier dynamics of the electron system after resonant optical excitation were also studied in bulk Ge in detail.[20] The efforts in the latter material culminated in the recent report of steady-state gain[21] and lasing[2] in highly doped tensilely strained Ge layers selectively grown on Si. Theoretical models of the underlying scattering processes between carriers and phonons were introduced and improved.[12,18,22–25] Extensive studies of the carrier dynamics have also been performed on compressively strained Ge quantum wells grown on Si. This material system is a particularly promising candidate for the Si integration of optical devices, as only recently direct gap electroluminescence at room temperature was observed.[26]

However, it remains unclear whether a strong coupling between electrons in the conduction band L valley and holes in the valence band Γ valley exists despite the diverse investigations of the carrier relaxation and cooling processes after optical excitation. In some models a common temperature for the electron and hole systems was assumed for reasons of simplicity,[12,23] yet in other approaches the coupling between holes and electrons was considered to be weak.[18,24] Experimental findings indirectly indicated a strongly coupled electron-hole system since the decay of the band-edge luminescence in intrinsic Ge was observed to be faster than that in p-doped Ge.[14] Recent photoluminescence studies on Ge quantum wells also suggest a strong coupling.[27] In this paper, we give direct experimental evidence for this process and, thereby, provide an answer to the enhanced free carrier absorption observed by Lange *et al.*[1]

II. METHODS

The indirect band structure of Ge makes it an ideal candidate to study the hole system's cooling dynamics using optical pump-probe spectroscopy independent of any distortions from conduction band states. Ge exhibits a local minimum at the Γ point; therefore, electrons can be optically excited in the conduction band Γ valley where they remain for a few hundreds of femtoseconds. Two processes dominate the dynamics of the electron system in the conduction band Γ valley during this time: On the one hand, Coulomb scattering rearranges the nonthermal carrier distribution toward a hot Fermi distribution and, on the other hand, the electrons are transferred into the lower lying L valleys by deformation potential scattering. The conduction band electrons experience only intervalley deformation potential scattering by acoustical and optical phonons, as the Fröhlich interaction is not present in nonpolar crystal structures. Additionally, intravalley deformation potential scattering is forbidden by symmetry for small k vectors around the conduction band Γ point.[28] Thermalization and intervalley scattering occur on timescales of tens to hundreds of femtoseconds. Thus, one can assume that no significant electron population is found in the conduction band Γ valley 1 ps after the excitation.[14,24,29] Consequently, the cooling dynamics of the electron system is invisible to

weak probe pulses, as there is no direct light-matter coupling between the valence band at the Γ point and the conduction band at the L point. In contrast to the electron system, the cooling dynamics of the hole system can still be accessed optically because the global minima of both the heavy and light hole valence bands are located at the Γ point. The relaxation of the hole system manifests itself by a clear bleaching at the lower-lying subbands' energies. Therefore, it is possible to deduce the underlying cooling dynamics of the hole system by monitoring the changes of the absorption as a function of the time delay between pump and probe pulses.

A. Experiment

Our measurements are performed in a fs-pump-supercontinuum-probe setup. The output laser pulses (800 nm, 1 mJ, 100 fs) of a 1 kHz Ti:sapphire amplifier system are split into the pump and probe beam. In the pump beam, an optical parametric amplifier is used to generate intense, 80 fs pump pulses with a tunable central wavelength ranging from 400 nm to 2200 nm. In the probe beam, self-phase modulation in a sapphire crystal is exploited to produce a white-light supercontinuum for the weak broadband pulses. After passing the sample, the probe light is spectrally dispersed in a 50 cm spectrograph and detected using a liquid-nitrogen-cooled (GaIn)As linear photodiode array yielding a spectral resolution of 1 nm. A delay stage with a maximum delay of 9 ns enables the time delay between pump and probe pulses with a time resolution of less than 50 fs. All experiments are performed at cryogenic temperatures on a multiple-quantum-well sample containing 50 compressively strained 10 nm thick Ge quantum wells between $Si_{(0.15)}/Ge_{(0.85)}$ barriers grown on Si. The band-edge absorption energy at 7 K is 0.96 eV. Further details on the sample were given earlier.[1] We performed excitation energy and photon fluency dependent measurements. Initially, the sample is excited slightly above the direct band gap at 0.965 eV. Then, the excitation is tuned to an energy in the continuum at 1.016 eV. In the last set of experiments the photon fluency was varied over two orders of magnitude for the 0.965 eV excitation.

III. RESULTS AND DISCUSSION

Typical experimental data for an excitation at 0.965 eV and of 4×10^{15} photons/cm^2 for different time delays between pump and probe are shown in Fig. 1. Initially a clear bleaching of the absorption at the lowest-lying subbands is observed. The bleaching decreases for increasing time delays until a major part of the absorption is recovered and a redshift accompanied by a broadening is found after 5 ps. Surprisingly, for later delay times the seemingly "recovered" absorption starts to show bleaching again and at 1 ns after the optical excitation the band-edge absorption almost reaches transparency. For later times, the system remains in that state for a few nanoseconds until the original absorption starts to recuperate. The recovery time of the system can only be estimated due to the limitation of the delay stage, yet it is well below the one millisecond corresponding to the repetition rate of the laser system.

We now focus on the indicated time scale of tens of ps; the ultrafast dynamics have been intensely studied and

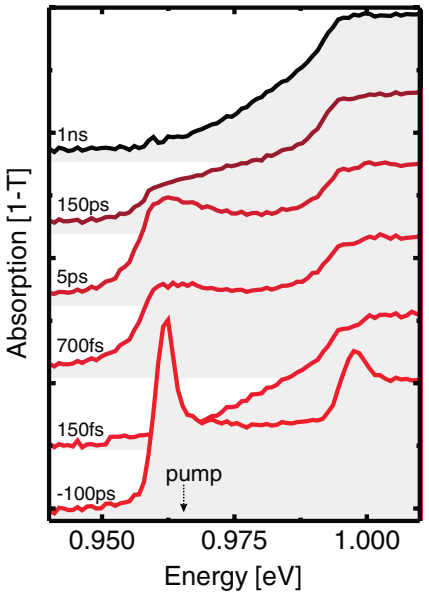

FIG. 1. (Color online) Typical time evolution of the Ge quantum well absorption around the direct band gap after optical excitation with 0.965 eV laser pulses. A clear ultrafast bleaching "recovers" after 1 ps and then starts to increase again on a picosecond timescale. The curves are shifted for clarity.

reported in the literature.[1,4,29] The detailed time evolution of the decreasing absorption for later delay times is depicted in Fig. 2(a). Initially, i.e., about 10 ps after excitation, we observe a slight bleaching and broadening of the first heavy-hole exciton resonance. For this time delay, all the excited electrons at the Γ valley have scattered into the L valley. With increasing time delay, a gradual decrease of the first heavy-hole subband absorption is observed. These relatively slow dynamics are explained by the relaxation of the hot hole system in the first heavy-hole valence band, cooling down toward the lattice temperature by deformation-potential scattering with optical and acoustical phonons. We exclude any spurious effects due to electrons excited into the barriers by two-photon absorption and their consecutive scattering into the quantum wells since the time evolution of the increasing bleaching is very slow compared to typical electron dynamics in germanium. And since no photoluminescence from the direct-gap transition is observed for high-energy excitation[30] it appears very unlikely that any electrons that might be in the barriers ever reach the Γ point. However, two-photon absorption contributes to the later bleaching signal at high photon fluencies starting around 5×10^{16} photons/cm^2 per pulse. Therefore, all experiments discussed here are performed an order of magnitude below those fluencies.

To support our explanation, we perform theoretical calculations modeling the absorption of an equilibrium carrier distribution at different carrier temperatures in a crystal lattice at lattice temperature $T_{lc} = 10$ K. The microscopic calculations of the absorption are based on the semiconductor Bloch equations including Coulomb effects on the Hartree-Fock level.[31] The band structure is modeled in a 30-band $\mathbf{k}\cdot\mathbf{p}$ model.[32] The experimental excitation and subsequent fast thermalization are described by the insertion of an initial

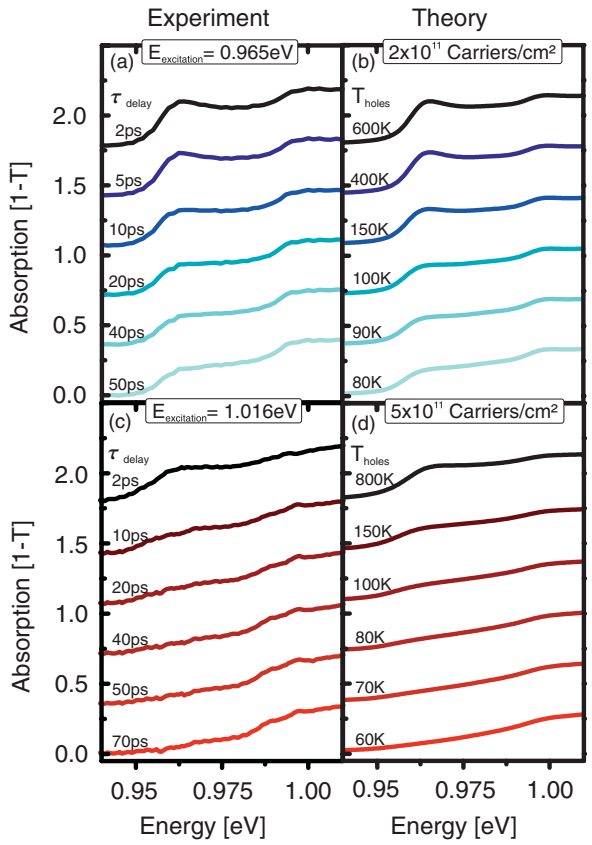

FIG. 2. (Color online) (a) Time evolution of the decreasing absorption for an excitation energy of 0.965 eV. (b) Calculated absorption for different hole temperatures showing very good agreement with the experiment shown in (a) and verifying the observation of a cooling hole system. (c) and (d) are similar to (a) and (b) and present results for a higher excitation energy (1.016 eV) showing no significant difference from the dynamics of (a), thus proving the minor contribution of the excess energy of the optical pulse to the heating of the holes in the monitored excess energy window. The curves are shifted for clarity.

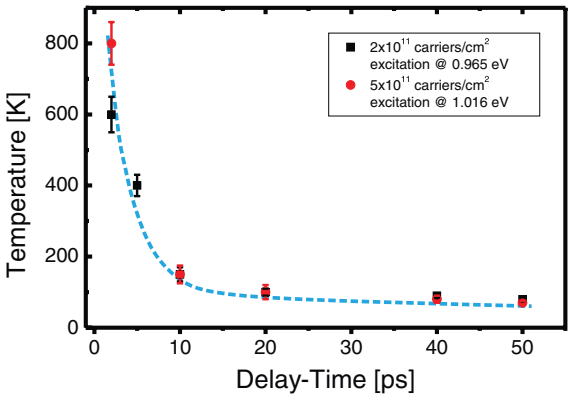

FIG. 3. (Color online) Theoretically extracted hole temperatures during the first 50 ps after the optical excitation are shown for two excitation energies, 0.965 eV and 1.016 eV, respectively. Near-resonant and far-above-resonant optical excitations with different carrier densities show similar hole temperatures and cooling characteristics. Hence, the source of the hole temperature has to be attributed to intrinsic scattering mechanisms. The dashed line is a guide to the eye.

thermal equilibrium carrier population at a carrier temperature T_c into the calculated band structure, the carriers being heated by the optical excitation with a surplus energy of 155 meV compared to the indirect band gap. The lattice, in contrast, is assumed to remain at a temperature $T_{lc} = 10$ K as is typically expected for experiments with pulsed, low-repetition-rate sources such as the 1 kHz amplifier system used here. We extract the system's hole temperatures and densities by varying these two parameters and comparing the calculated spectral line shape to the measured spectra for the different time delays. Typical spectra are shown in Fig. 2(b). Note that no linear relationship between photon fluency and calculated carrier density exists due to nonlinearities, such as Pauli blocking during the excitation pulse.

The simulated absorption spectra yield very good agreement with the experiment for all conditions. Therefore, we conclude that we observe a cooling hole system where we can quantitatively determine its carrier density and temperature dynamics. Strikingly, we encounter temperatures of up to 600 K for the hole system in the first few picoseconds after the optical excitation, in contrast to the excess energy of the optical excited holes in the valence band with at most 10 meV (about 116 K). Consequently, the excess energy of the optical excitation with respect to the band extrema at the Γ points alone cannot explain the hot hole system. The same holds true for experimental results with higher excitation energy shown in Fig. 2(c), with the corresponding theoretical simulations in Fig. 2(d). Here, the excess energy of 56 meV corresponds to a maximum temperature of 650 K. Nevertheless, the simulation yields temperatures of 800 K 2 ps after excitation. Pump-probe experiments where the pump pulse energy was tuned below the direct band gap energy reveal that indirect absorption by emission or absorption of phonons is at least an order of magnitude weaker; therefore, it can be neglected. Hence, possible scattering or heating processes have to be considered in order to explain the apparent energy gain of the holes. This heating process occurs on the ultrafast time scale within the thermalization time of the electron system in the L valley, as only a small residual bleaching of the hot hole system and a redshift of the electrons in the L valley are observed 700 fs after excitation. The energy difference of the conduction band Γ point and the lower-lying conduction band L valleys in the analyzed system amounts to 150 meV, corresponding to a maximum temperature of 1740 K. Considering an energy transfer from electrons to holes so that both systems share the same temperature leads us to a maximum temperature of 870 K. We thus conclude that the hot electron system heats the hole system. As the thermalization time of the electrons in the L valley is of the order of the heating time of the holes, and the energy difference of Γ and L valley corresponds very well to the determined hole system temperatures.

The extracted cooling curves for both excitation energies are depicted in Fig. 3. Both excitation energies yield similar results in respect to the temperature dynamics of the hole system. The observed time dependence is comparable to the hole cooling times observed by Woerner *et al.*[15] In the first

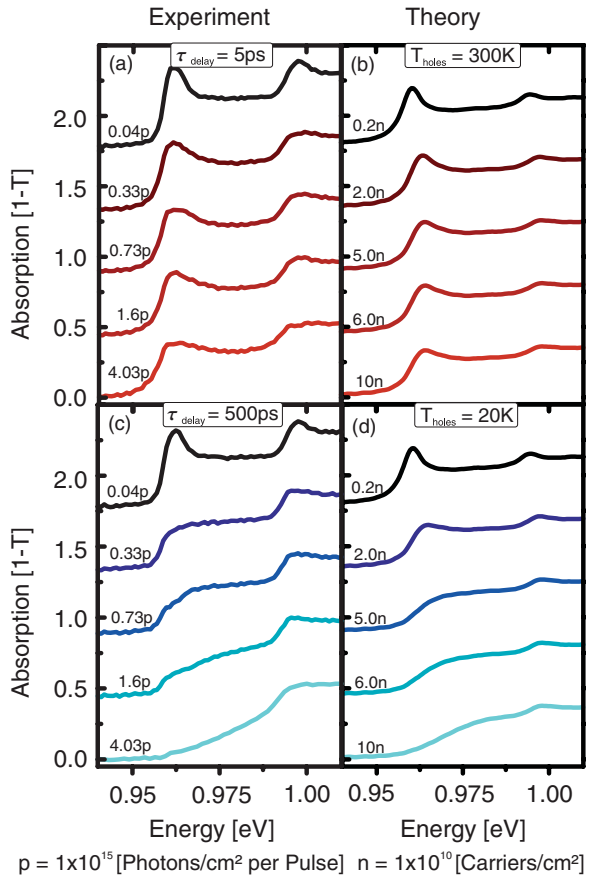

FIG. 4. (Color online) Influence of the carrier density on the heating and cooling of the hole system by an excitation energy of 0.965 eV. p is the scaling factor for the different photon fluencies and n is the scaling factor for the different carrier densities. (a) Experimental and (b) calculated absorption spectra for different excitation intensities and a time delay of 5 ps showing no change in the hole system temperature. (c) and (d) are the same as (a) and (b) but for a time delay of 500 ps demonstrating that the cooled hole systems have reached equal temperatures but the pump probe signals show different strengths of bleaching due to different carrier densities. The curves are shifted for clarity.

few picoseconds deformation potential scattering by optical phonons dominates the cooling and is responsible for the rapid decrease of the temperature until it is suppressed by Pauli-blocking effects. Then deformation potential scattering by acoustical phonons becomes the main scattering mechanism cooling the carrier system to the lattice temperature on a nanosecond time scale.

In the last set of experiments, the intensity dependence is investigated. The absorption spectra for the delay times of 5 ps and 500 ps for various excitation intensities covering more than two orders of magnitude are shown in Fig. 4(a); corresponding theoretical calculations are displayed in Fig. 4(b). A strong difference in the absorption spectra is observed for the different excitation intensities. Yet, the theoretical calculations reveal that only the carrier density of the hole system is changed and not the temperature. Consequently, higher carrier densities do not affect the heating mechanism of the hole system significantly. Previous works pointed out that screening effects of the deformation potential intervalley scattering do not play a role in this system as the screening distance is approximately the interparticle distance, which is still smaller than the lattice constant.[14,19] Thus, the intervalley scattering rate is not altered for higher carrier densities resulting in similar carrier distributions in the Γ and L valleys for all intensities. Furthermore, in contrast to GaAs, where the Fröhlich interaction involves phonons over a narrow energy range, in Ge the deformation potential involves phonons over a broader energy range; hence no hot phonon effect was observed here.[12] Considering that higher nonequilibrium phonon populations do not lead to higher reabsorption rates of phonons in the hole system in Ge, we conclude that phonons emitted by the thermalizing and scattering electron system are not the driving force of the hole system's heating process. Instead, we attribute this to direct Coulomb-potential scattering between electrons and holes. An efficient Coulomb scattering process between electrons and holes could be explained by their similar effective masses.[12,14,19] However, further microscopic calculations are necessary to support this thesis.

Finally, we discuss the implication of this heating mechanism for the enhanced free carrier absorption after near-resonant excitation reported in Lange *et al.*[1] It qualitatively explains the observed difference of the free carrier absorption between long (picosecond) and ultrashort (femtosecond) excitation. In the experiments reported here, transient gain was only observed for femtosecond excitation. An increased free carrier absorption arising below the band gap a few femtoseconds after the excitation competes with the reported gain, eventually dominating it. For picosecond excitation, no transient gain at all was observed and the shape and time evolution of the free carrier absorption is distinctively different. Leung and Scully[10] showed that the free carrier absorption in Ge is strongly dependent on the hole system temperature. Therefore, the strong free carrier absorption after resonant optical excitation can be attributed to the heated hole system. Furthermore, as the cooling dynamics appear on a picosecond time scale, excitation with femtosecond and picosecond pump pulses leads to different hole temperatures which again lead to different absorption characteristics.

IV. SUMMARY AND CONCLUSIONS

We have experimentally observed a strong heating of the hole system after nearly resonant optical excitation due to the energy from the thermalizing electron system in the L valley. The experimental findings are supported by theoretical simulations clearly identifying the cooling of the hole system. This heating process is assigned to an efficient Coulomb scattering, resulting from similar effective masses of electrons in the conduction band L valleys and holes in the valence band Γ valley. The heated hole system gives an explanation for the enhanced free carrier absorption after resonant optical excitation observed by Lange *et al.*[1] At last, in optical applications of Ge such as saturable absorbers and optical modulators this effect needs to be considered and might lead to consequences in the practicability of Ge as a material system for fast optical switching.

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