Femtosecond Carrier Dynamics in GaAs Far from Equilibrium

Alfred Leitenstorfer, Cornelius Fürst, Alfred Laubereau, and Wolfgang Kaiser
Physik Department E 11, Technische Universität München, D-85748 Garching, Germany

Günther Tränkle and Günter Weimann
Walter-Schottky-Institut, Technische Universität München, D-85748 Garching, Germany
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The dynamics of optically generated carriers in GaAs is investigated measuring transmission changes with ultrafast time and high spectral resolution. A novel two-color 15 fs Ti:sapphire laser system allows the observation of the femtosecond kinetics of energetic carrier distributions at excitation densities as low as 10^{15} cm^{-3}. For the first time, LO phonon emission of highly excited nonequilibrium electrons is directly monitored. The contribution of the hole distribution to the data and the influence of carrier-carrier scattering is studied.

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A detailed understanding of the basic scattering mechanisms of highly energetic carriers in semiconductors is essential for various problems in modern physics and technology. Experiments investigating these effects provide valuable information for subjects ranging from the development of high speed electronic devices to fundamental questions in many-body and plasma physics. Optical spectroscopy with ultrashort laser pulses is a major tool for the observation of hot carrier dynamics, since the strong coupling of elementary excitations, a characteristic property of solids, leads to a rapid relaxation typically occurring on a subpicosecond time scale. In intrinsic semiconductors, there are two major classes of interaction mechanisms responsible for this ultrafast temporal evolution: carrier-carrier (cc) and carrier-phonon (cp) scattering. The best insight into these fundamental processes is provided by experiments where the different contributions to the kinetics can be clearly separated. However, time and energy resolved data under conditions where cp scattering is the dominant effect and energy redistribution by cc collisions is negligible are difficult to obtain because of the limited sensitivity of ultrafast measurements at very low carrier concentrations.

Studies of hot carrier distributions with femtosecond time resolution have been carried out with two well-established techniques: time and energy resolved transient absorption measurements [1–4] and luminescence up-conversion [5,6]. It turned out that these previous experiments required excitation densities typically $>5 \times 10^{16}$ cm^{-3}. In this regime, the rapid cc collisions dominate leading to a thermalization of the carriers on a 100 fs time scale. At densities around 10^{14} cm^{-3}, nonequilibrium dynamics of hot electrons has been studied employing a sensitive streak camera with a limited time resolution of a few picoseconds [7]. Energy exchange of the hot carriers with the crystal lattice could not be observed independently in these earlier investigations. In particular, energetic structure originating from the dominant process of electron-phonon scattering in polar semiconductors, the Fröhlich interaction with longitudinal optical (LO) phonons has not been time resolved. We recall that LO phonon scattering is a major intrinsic source of electric resistance in most direct gap materials. Nonequilibrium features of highly energetic electron distributions due to LO phonon emission in GaAs and InP have been seen in spectrally resolved hot luminescence studies with cw, ps, and fs excitation [8–12]. Although time integrated, these experiments give information on cc scattering times [11], band structure [12], or the influence of phase relaxation on the generation process [10].

In this Letter, we present transient absorption changes in GaAs measured with the help of a novel two-color Ti:sapphire laser [13]. The high repetition rate (76 MHz) together with the spectral tunability of our system enables us to observe for the first time the femtosecond kinetics of the energetic distributions of hot carriers at excitation densities as low as 1.5×10^{15} cm^{-3}. In addition, time resolution is pushed on to the 10 fs regime. In our experiments, a 500 nm thick layer of high purity GaAs grown by molecular beam epitaxy is studied [14]. The sample is antireflection coated and held at a lattice temperature of $T_L = 15$ K. Free electron-hole pairs are excited above the band gap ($E_{\mathrm{gap}} = 1.52$ eV) with transform-limited Gaussian pump pulses of a duration $t_p = 80$ fs and a central photon energy of 1.63 eV. Exciting GaAs with this energy generates electrons out of the heavy hole and light hole bands. Momentum conservation results in a sharing of the excess photon energy between the particles according to the reciprocal ratio of their effective masses: The electrons involved in the transition from the heavy hole to the conduction band posses an initial excess energy of 10 meV, substantially above the threshold for LO phonon emission (phonon energy $\hbar\omega_{\mathrm{LO}} = 36$ meV). The corresponding heavy hole distribution is centered around an energy of 12 meV. Excitation out of the light hole band yields electrons of an excess energy of 65 meV and light

holes of 47 meV. The differential transmission $\Delta T/T$ induced in the sample is monitored with weak probe pulses of duration $t_p = 15$ fs and centered at photon energies of 1.57 eV. Pump and probe pulse trains are synchronized to better than 2 fs [13] with linear polarization adjusted perpendicular to each other. Energy information is obtained by analyzing the spectrum of the transmitted probed pulses applying a double monochromator with the spectral resolution set to 4 meV. The large bandwidth of the short pulses allows probing in an energy interval of approximately 200 meV.

In Fig. 1 we present energy and time resolved transmission changes recorded after excitation of 1.5×10^{15} electron-hole pairs per cm^3 at four delay times of 40, 100, 200, and 500 fs. The signals are dominated by the bleaching of the transition from the heavy hole to the conduction band due to the photoexcited electron population. At a delay time of 40 fs with respect to the maximum of the pump pulse ($t_p = 80$ fs), carrier generation is almost completed. In the corresponding spectrum [Fig. 1(a)] the most prominent peak appears at a photon energy of 1.62 eV (heavy hole), representing the electrons excited from the heavy hole band. A less pronounced maximum is situated near 1.58 eV (light hole), being due to the electrons originating from the light hole band. The relative magnitude of the two peaks is in good agreement with the ratio of 5:2 which is expected theoretically for excitations out of the two valence bands [15]. After 100 fs [Fig. 1(b)] the maximum at 1.62 eV has clearly decreased, while the peak at 1.58 eV increased. This effect is explained by electrons excited from the heavy hole band which have emitted one LO phonon of an energy of 36 meV, thus scattering in the energy region originally populated by the light hole electrons. Of special interest is the observation of a third maximum (−2LO) arising near 1.53 eV at a time delay of 200 fs [Fig. 1(c)]. This peak corresponds to electrons originating from the light and heavy hole bands which have emitted one or two LO phonons, respectively. At a time delay of 500 fs [Fig. 1(d)] most electrons have relaxed to the bottom of the conduction band where no LO phonon emission is possible.

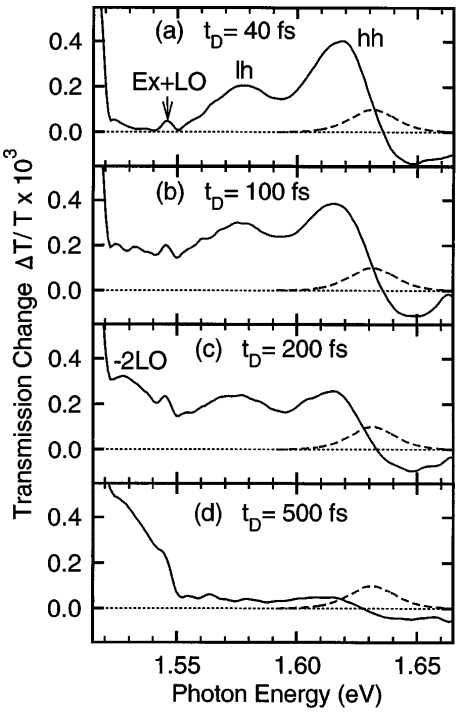

FIG. 1. (a)–(d) Spectrally resolved transmission changes $\Delta T/T$ measured for four different probe delays t_D at an excitation density of $N = 1.5 \times 10^{15}\,cm^{-3}$ and at a lattice temperature of $T_L = 15$ K. Dashed lines: spectrum of the excitation pulse.

The steep increase in transmission observed near 1.52 eV is caused by the screening of exciton states (higher than 1*s*) by the generated charge carriers. Screening of the electron-hole attraction strongly reduces the exciton's oscillator strength leading to a substantial decrease of the absorption. At all time delays, we observe a weak narrow band near 1.55 eV (Ex + LO), at the energy of one optical phonon above the 1*s* exciton in our sample. We attribute this novel feature to the bleaching of a two quantum absorption process, where one photon creates an exciton together with an optical phonon. This line has not been seen, to our knowledge, in previous linear absorption spectra since the two quantum process is approximately 2 orders of magnitude weaker than the direct transitions prevailing above the band gap. The peak appears more pronounced in our measurements because of the high nonlinearity of the excitonic absorption.

Another interesting phenomenon is found around the probing energy of 1.65 eV. In sharp contrast to the expected bleaching by the carrier population, a pronounced induced absorption appears above the maximum of the pump spectrum: The nonequilibrium distribution of electrons causes an increase of the Coulomb enhancement factor of the absorption at its high energy side [16]. This many-body effect is closely related to the Fermi edge singularities in metals and highly doped semiconductors [2,17]. The simultaneous slight redshift of the bleaching peak has previously been discussed at much higher excitation densities [2,18]. We emphasize that at our low carrier concentrations the induced absorption still exists strongly at delay times substantially longer than the duration of the excitation pulses [Figs. 1(b) and 1(c)].

In order to demonstrate the increasing influence of cc scattering we present in Fig. 2 four spectra obtained at a 1 order of magnitude higher density of $2 \times 10^{16}\,cm^{-3}$. All other parameters remain equal to the data in Fig. 1. In this regime, the rapid dephasing due to cc collisions results in a broadening of carrier generation in k space [10]. In addition, energy exchange between the carriers is also more efficient. Both effects contribute to the less pronounced structure found in the spectra depicted in Fig. 2. However, the time resolved measurements clearly

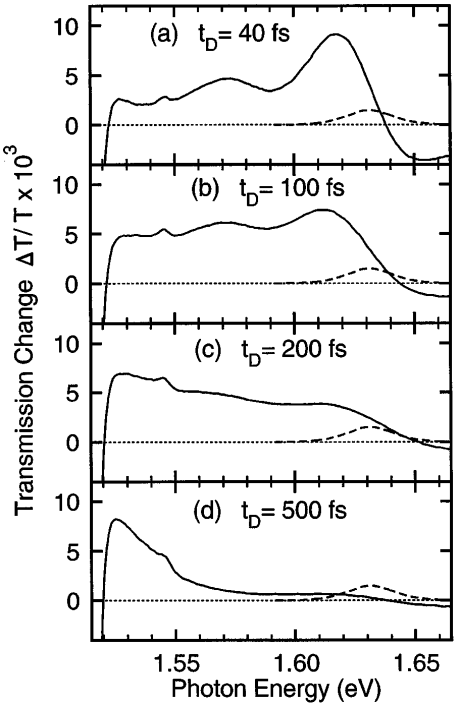

FIG. 2. (a)–(d) Spectrally resolved transmission changes $\Delta T/T$ at an excitation density of $N = 2 \times 10^{16}$ cm^{-6} for the same delay times and for equal parameters as in Fig. 1.

show that the initial energy relaxation of the electrons is still strongly affected by the relatively slow LO phonon emission. As a consequence, the electrons have not yet relaxed into a thermal equilibrium distribution after 200 fs [Fig. 2(c)]. This behavior is completely different to the kinetics found at densities above 10^{17} cm^{-3}, where the effective energy exchange among carriers leads to an ultrafast thermalization almost completed after 100 fs [4,5]. The spectra taken at a concentration of 2×10^{16} cm^{-3} show a strong decrease in transmission at 1.52 eV indicating a larger influence of renormalization effects which lead to an enhanced absorption below the original gap energy.

At the late delay times of 500 fs [Figs. 1(d) and 2(d)], for both excitation densities a small, nearly constant, bleaching between 1.56 and 1.62 eV is observed. The hot electrons cannot account for this finding, since after 500 fs the occupation of electrons in states high in the conduction band is negligible. We note that also holes can contribute to the transmission changes seen in our experiment. While the light holes should relax rapidly, the heavy holes generated by the pump photons are below the threshold for LO phonon emission and thus might bleach transitions around 1.63 eV for longer times. Recently, it was demonstrated that for comparable energies of electron and heavy hole distributions the transmission changes observed probing high above the band gap are dominated by the holes [19].

To obtain more quantitative information on the dynamics of electrons and holes, we performed a series of time resolved measurements with a probing energy of 1.62 eV, corresponding to the maximum of the most prominent initial bleaching peak. The temporal evolution of $\Delta T/T$ for the two densities discussed above is shown in the semilogarithmic plots of Fig. 3. The data were corrected for a weak negative background due to band-gap renormalization [19] taken from our data at a delay time of several picoseconds.

At a density of 1.5×10^{15} cm^{-3} [Fig. 3(a)], we find a kinetics with a biexponential behavior: The fast initial dynamics shows a decay time $\tau_1 = 220 \pm 20$ fs. As expected, this finding is in good agreement with LO phonon emission times of hot electrons in GaAs reported previously [20,21]. Additional time resolved data give the same value of τ_1 independent of density below 3×10^{15} cm^{-3}, indicating that energy relaxation by cc scattering is inefficient compared to LO phonon emission. At densities above approximately 3×10^{15} cm^{-3}, τ_1 is found to become significantly faster. The growing contribution of cc scattering accelerates the energy redistribution of the electrons [e.g., $\tau_1 = 185 \pm 15$ fs for $N = 2 \times 10^{16}$ cm^{-3}, Fig. 3(b)]. These results quantitatively agree with theoretical predictions for the scattering rates of nonthermal carriers in GaAs, including full dynamical screening of the cc interaction [22]: For a Gaussian electron distribution of an energetic width of

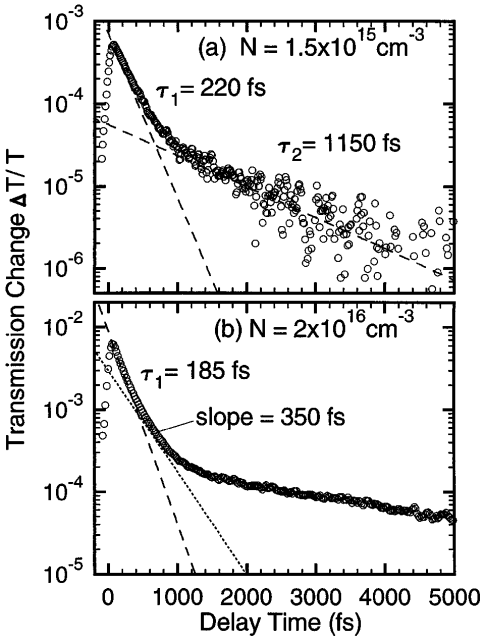

FIG. 3. Transmission changes $\Delta T/T$ at a probing energy of 1.62 eV vs delay times for two different carrier concentrations N. (a) The dashed lines represent two components with time constants τ_1 and τ_2 of a biexponential fit to the data. (b) A monoexponential fit to the initial decay time τ_1 is indicated by the dashed line. A slower component is suggested by the reduced slope around $t_D = 700$ fs (dotted line).

20 meV centered 100 meV above the band edge, an effective electron-electron scattering rate of 1 ps^{-1} is calculated at a density of 2×10^{16} cm^{-3}. Combined with an LO phonon emission rate of 4.7 ps^{-1} [22], an initial decay time of the electron distribution around $\tau_1 =$ 175 fs is expected. Assuming an $N^{1/2}$ dependence for the energy relaxation via cc collisions [23], we estimate the corresponding scattering rate to be approximately 0.2 ps^{-1} at a density of 1×10^{15} cm^{-3}. As observed experimentally for $N < 3 \times 10^{15}$ cm^{-3}, this small cc scattering contribution is below the accuracy of our measurement.

The second time constant τ_2 in Fig. 3(a) may be attributed to the slow relaxation of heavy holes. In contrast to the electrons, hole relaxation is more strongly dependent on carrier density in the range of our investigation, i.e., between 5×10^{14} and 5×10^{16} cm^{-3}. We recall that the heavy holes cannot emit LO phonons on account of their low excess energy. However, the high density of final states for cc collisions within the heavy hole band leads to higher rates for hole-hole scattering compared to electron-electron scattering. From a series of measurements with increasing carrier concentrations, we can trace the time constant $\tau_2 = 1150 \pm 200$ fs observed at 1.5×10^{15} cm^{-3} [Fig. 3(a)] changing into a slope of approximately 350 fs obtained at the higher density of 2×10^{16} cm^{-3} around a delay time of 700 fs [dotted line in Fig. 3(b)]. We note that a densities above 3×10^{15} cm^{-3} the data cannot be fitted with two exponential time constants within the first picosecond after excitation. This finding can be related to the increasing influence of the cc scattering rates. The latter are themselves strongly time dependent due to the rapidly varying carrier distributions [24]. The origin of the weakly pronounced picosecond dynamics observed at higher densities [for time delays $t_D > 1500$ fs in Fig. 3(d)] is complex in nature. Processes such as heavy hole cooling and/or diffusion effects within the thin sample contribute to the late signal decay.

In conclusion, we have presented for the first time an energy resolved study on carrier dynamics in a regime where energy exchange with the lattice is fast compared to carrier-carrier scattering. LO phonon emission is seen to dominate the relaxation of electrons at densities around 10^{15} cm^{-3}. Increasing the excitation density by 1 order of magnitude results in an additional broadening of the observed features by the cc interaction while some structure originating from LO phonon emission is still discernible. The kinetics of the heavy holes is found to be strongly dependent on carrier concentration.

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