Direct Subpicosecond Measurement of Carrier Mobility of Photoexcited Electrons in Gallium Arsenide

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The mobility of hot electrons in gallium arsenide has been measured with subpicosecond time resolution, following injection by 2-eV femtosecond pulses. The mobility immediately after injection was measured to be less than 500 $cm^2/V \cdot s$, indicating efficient electron transfer into the satellite L valleys. The electron mobility then rose to a quasiequilibrium value of 4200 $cm^2/V \cdot s$, limited by electron-hole scattering. The rise time was observed to be between 1.8 and 3.2 ps depending on injected carrier density.

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Measurements of the dynamics of hot electrons in semiconductors are extremely important for advancing our knowledge of the physics of nonequilibrium phenomena in these materials. They yield information about carrier-phonon and carrier-carrier interactions, being of fundamental interest in semiconductor physics. On the other hand, knowledge of hot-electron dynamics is crucial for the understanding of the physical basis of high-field phenomena like velocity overshoot, ballistic transport, and resonant tunneling. Previous measurements have used either electronic or optical techniques to measure hot-electron relaxation. In the first case, direct electronic measurements of the drift velocity have been made by microwave and time-of-flight techniques.[1,2] The time resolution of these approaches is not, however, sufficient to resolve directly the relaxation processes occurring on a time scale varying from tens of femtoseconds to several picoseconds. All-optical techniques which measure transmission or luminescence[3] can readily be applied to the observation of carrier relaxation phenomena and have subpicosecond time resolution.[4-6] Unfortunately, optical probes do not measure electronic transport parameters, which can only be inferred indirectly from the optical properties with exact knowledge of the band structure.

In this paper, we describe an experiment which combines the speed advantage of optical excitation with direct subpicosecond electrical measurements of the drift mobility. The use of subpicosecond electrical pulses which are synchronized to the optical excitation pulse has made possible the first direct measurements of nonequilibrium electronic transport on a subpicosecond time scale. In addition, the measurement does not require electrodes on the sample and can be calibrated to give absolute values of the mobility. Our results give direct experimental evidence for relaxation of hot electrons from the L valley to the central Γ valley.

Our experiment uses femtosecond electrical pulses generated by optical rectification of femtosecond laser pulses in the electro-optic crystal $LiTaO_3$.[7] These pulses consist of a single cycle of electromagnetic radiation with a duration as short as 300 fs and a spectrum that peaks in the far-infrared range of the electromagnetic spectrum ($\simeq 1$ THz). A schematic of the experimental setup is depicted in Fig. 1. Optical pulses of 60 fs duration are generated in a colliding-pulse mode-locked (CPM) laser ($\lambda = 625$ nm) and amplified by a copper vapor laser to $\simeq 1$ μJ energy per pulse at 8-kHz repetition rate. A small portion of this beam ($\simeq 4\%$) is split off to derive two beams for the generation and probing of the femtosecond electrical pulse. Both beams are focused in parallel to spot sizes of 8 μm diameter into the 1-mm-thick $LiTaO_3$ crystal, separated by 20 μm (Fig. 1). The electrical pulse evolves from the generating beam in the form of a conical Cherenkov shock wave.[7] The second, much weaker, probe beam serves to measure the propagating electrical pulse via electro-optic sampling. The complete waveform can be mapped out by a change of the time delay t_{probe} between generating and probing beams.

The GaAs sample is optically contacted to a polished crystal face of the $LiTaO_3$ crystal. The intrinsic sample

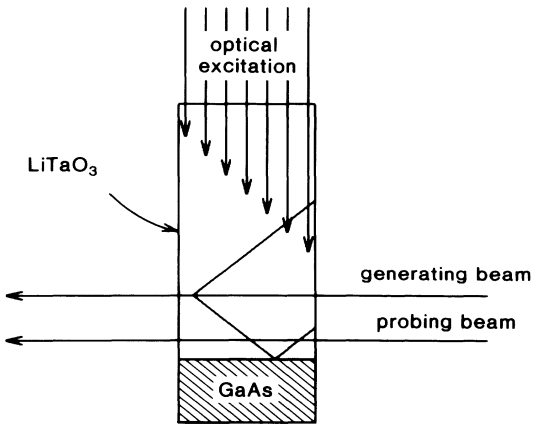

FIG. 1. Schematic of the experimental arrangement.

is grown by molecular-beam epitaxy on a semi-insulating GaAs substrate and consists of a 9300-Å-thick buffer layer of GaAs followed by a 1900-Å-thick $Al_{0.52}Ga_{0.48}As$ layer, a 4600-Å-thick GaAs, and a 105-Å-thick $Al_{0.33}Ga_{0.67}As$ cap layer. The conical wave of the femtosecond electrical pulse sweeps across the surface of the sample, is reflected, and crosses the probe beam again after a certain delay time. From the face of the $LiTaO_3$ crystal opposite the GaAs crystal, the remaining 96% of the 60-fs optical pulse illuminates the sample after passing through a transmission echelon. The echelon provides a tilted wave front of the optical pumping beam in synchronism with the conical wave of the electrical pulse. The optical excitation pulse creates carriers with a density between 5×10^{17} and 1.2×10^{19} cm^{-3} within the 2500-Å absorption depth with an excess energy of 500 meV for the electrons and 70 meV for the holes.

For each delay T_{ex} of the optical carrier excitation pulse, the reflected electrical pulse is now recorded. A series of traces obtained this way is plotted in Fig. 2. Time zero of the delay T_{ex} and the time resolution of the plot is determined by a cross-correlation experiment of the optical excitation pulse and the optical probe pulse using the third-order nonlinear susceptibility $\chi^{(3)}$ of the $LiTaO_3$ crystal and has a FWHM of 290 fs. The reflected electrical pulse shape plotted in the rear of the figure is recorded without optical excitation and represents the dielectric response of the GaAs crystal without free carriers. As we go towards the front of the set, the phase as well as the amplitude changes dramatically. In particular, the phase changes by 180° as expected for an insulator-to-conductor transition.

The electric field of the reflected femtosecond electrical pulse E_r can be derived from the boundary condition at the thin conducting layer at the interface between $LiTaO_3$ and GaAs:

$$E_r(t)=r_0E_i(t)-Y^{-1}J_s(t), \tag{1}$$

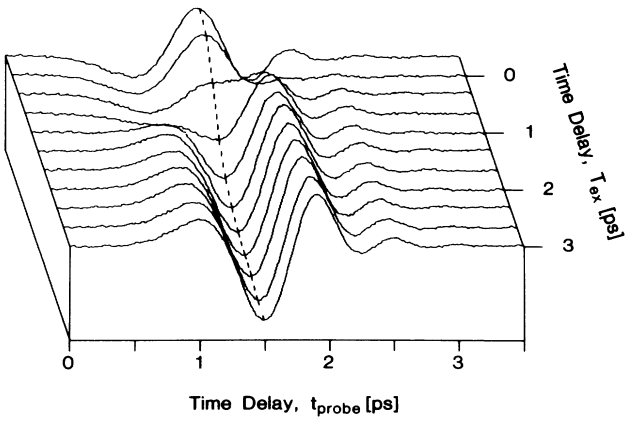

FIG. 2. Reflected electrical pulses for different settings of the optical pump delay T_{ex}. The dashed line marks the probe delay at which the data of Fig. 3 were taken.

where r_0 is the reflection coefficient of the interface without the presence of free carriers, J_s the sheet current in the thin conducting layer, and $Y=(\epsilon_0/\mu_0)^{1/2}\times(\epsilon_1^{1/2}\cos\theta_i+\epsilon_2^{1/2}\cos\theta_t)$ with θ_i, θ_t being the angles of incidence and transmission and ϵ_1,ϵ_2 the dielectric constants of $LiTaO_3$ and GaAs, respectively. As can be seen from Fig. 2, only for the short time of carrier injection around $T_{ex}=0$ does the phase of the reflected electrical pulse change; it remains nearly constant during the subsequent cooling of the hot carriers. This means that the sheet current J_s follows the electric field $E(t)$ directly without phase change, so that Ohm's law is instantaneous in time: $J_s(t)=\sigma(t)E(t)$. Ohm's law is valid here even for hot carriers, since the electric field of the probing pulse is small ($\simeq$100 V/cm). $E(t)=E_i(t)+E_r(t)$ is the driving field of the carriers in the conducting sheet. We then gain an expression for the reflected electric field E_r:

$$E_r=\left\{r_0-(1+r_0)\frac{\sigma_s}{Y+\sigma_s}\right\}E_i=r(t)E_i(t). \tag{2}$$

The expression in braces is the reflection coefficient $r(t)$. With the reflection coefficient known, the conductivity $\sigma_s=Y(r_0-r)/(1+r)$ can be obtained. Our technique provides at the same time a calibration of the injected carrier density by comparison of the reflectivity after complete cooling with the reflectivity estimated from a Drude model.[8] Therefore, except for the short time ($\simeq$290 fs) during which the carriers are injected, the mobility of the electron gas can be derived from the ex-

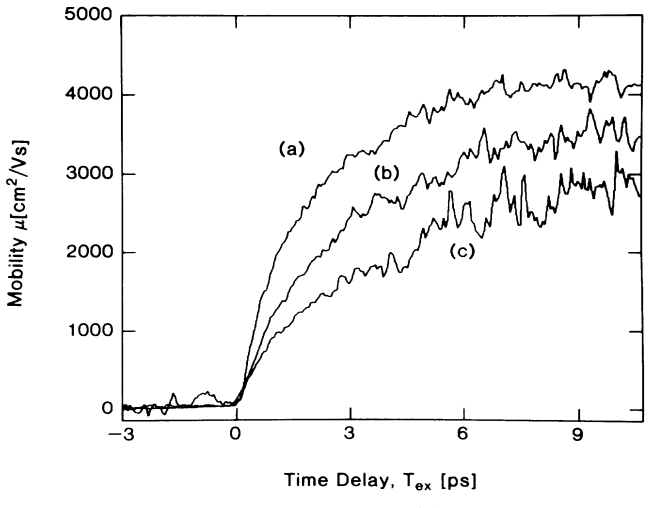

FIG. 3. The right-hand side of Eq. (3) obtained from the reflection coefficient $r(t)$ as function of delay time T_{ex} after carrier injection. For $T_{ex}>0.3$ ps, this parameter is identical to the carrier mobility μ. The traces a–c correspond to injected carrier densities of 5×10^{17}, 5×10^{18}, and 1.2×10^{19} cm^{-3}, respectively.

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$$\mu(t)=(\sigma_s/n_s e)[1+r(t)], \quad (3)$$

where n_s is the sheet carrier density and e the electronic charge. The mobility $\mu(t)$ obtained according to Eq. (3) from the reflection coefficient at fixed probe delay (marked by a dashed line in Fig. 2) is plotted in Fig. 3. Since the mobility for the holes is much lower than for the electrons, the data shown in Fig. 3 represent essentially the electron mobility.

The change of the mobility as a function of delay time T_{ex} of the optical excitation pulse is shown for three different injected carrier densities in Fig. 3. The transient mobility data of Fig. 3 show three extremely interesting features:

(i) The electron mobility immediately after injection is < 500 cm^2/V·s. This low value cannot be explained exclusively by the hot-electron distribution in the central Γ valley. For excess energies above the intervalley separation of 0.31 eV, intervalley scattering must be taken into account. At 500 meV excess energy, the intervalley scattering rates are extremely high ($\tau_{\Gamma L} \simeq 40$ fs[9]), so that a substantial fraction of the injected electrons will scatter into the L valley, having a very low mobility of $\simeq 250$ cm^2/V·s.[10] Monte Carlo simulations show that under similar conditions, almost 80% of the injected electrons have scattered into the L valley within 200 fs.[11] Therefore, the measured rise time of the mobility can be attributed largely to the intervalley transfer from the low-mobility L valley back to the high-mobility Γ valley. These estimates are corroborated by the low initial mobility measured directly after the carrier injection. The measured time constant of $\simeq 2$ ps for this transfer is much slower than the initial $\Gamma \rightarrow L$ intervalley scattering time, as expected from the difference in the densities of the final states. The fraction of the electrons remaining in the central valley evolves rapidly (within < 100 fs) into a Boltzmann distribution with an electron temperature $T_e \gg T_L$. This distribution cools down to the lattice temperature T_L by LO-phonon emission with a time constant of several picoseconds. Since the mobility changes associated with this cooling process are small compared to the intervalley transfer,[1] the contribution to our mobility data is small.

(ii) The maximum mobility for long delay times (> 10 ps), when the electron temperature has reached the lattice temperature, decreases from 4200 cm^2/V·s at a carrier density of 5×10^{17} cm^{-3} to 3500 cm^2/V·s at $n=5\times10^{18}$ cm^{-3} and to 2900 cm^2/V·s at $n=1.2\times10^{19}$ cm^{-3} and even at the lowest carrier density is much less than the maximum electron mobility of $\simeq 7000$ cm^2/V·s of the sample grown by molecular-beam epitaxy. Because of the sandwiching of the GaAs between two AlGaAs layers, surface scattering should be negligible. Polar optical scattering actually decreases with increasing electron density and yields mobilities at least two times higher than observed in our measurements.[12] However, photoexcitation injects both electrons *and* holes of equal density. Since the holes are much heavier than the electrons ($m_e=0.067m_0$, $m_h=0.57m_0$), they act as nearly elastic scattering centers for the electrons, thereby reducing the electron mobility.[13] At 300 K the electron-hole scattering can be adequately described by the equation for ionized-impurity scattering with the electron mass replaced by the reduced mass.[14] The agreement of the calculated mobilities with our experimental data is within 20% for the lowest carrier density, but predicts too low mobilities for the higher electron densities. This is probably due to the electron gas not being completely nondegenerate at the highest densities. Electron-electron scattering, however, is not effective in reducing the carrier mobility, since the average momentum of the electron gas is conserved during collisions.

(iii) The mobility rise time is strongly affected by the number of injected carriers. A fit to the data in Fig. 3 reveals an exponential time constant of 1.8 ps at a carrier density of $n=5\times10^{17}$ cm^{-3}, which increases to 2.6 ps at $n=5\times10^{18}$ cm^{-3} and to 3.2 ps at $n=1.2\times10^{19}$ cm^{-3}. We believe that the dependence of the mobility rise time on carrier density observed in our experiment is due to the generation of nonequilibrium LO phonons during the cooling process. This effect has been shown to decrease the cooling rate of a highly excited semiconductor plasma considerably[3,15,16] and will also reduce the intervalley transfer rate, since LO phonons are emitted during this process.

In conclusion, we have determined the mobility of hot photoinjected carriers in GaAs directly with < 300-fs time resolution using femtosecond electrical pulses. We find very low electron mobilities (< 500 cm^2/V·s) immediately after injection as a result of efficient intervalley scattering into the L valley. The mobility then increases with a time constant between 1.4 and 3.2 ps for injected carrier densities ranging between 5×10^{17} and 1.2×10^{19} cm^{-3}, respectively. The measured time constant is largely determined by the intervalley relaxation process from the low-mobility L valley back into the high-mobility central valley. To our knowledge, this is the first direct measurement of this intervalley relaxation process in GaAs. We find that the equilibrium mobility (after > 10 ps) decreases with increasing density and is limited by electron-hole scattering.

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