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# Intervalley scattering in GaAs and InP probed by pulsed far-infrared transmission spectroscopy

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The dynamics of photoexcited electrons in GaAs and InP were studied using the transmission of 200-fs pulses of far-infrared radiation in the spectral range 15–100 $cm^{-1}$. Kinetic traces of the infrared transmission as a function of delay between optical excitation and infrared probe show a probe-limited decrease in transmission followed by a more gradual (0.7–2 ps) drop to a steady value, consistent with the slow return of electrons from high-mass satellite valleys. Infrared transmission spectra, analyzed in the context of a Drude model, reveal density-dependent electron mobilities 3–4 times below equilibrium *n*-doped values. Electron-hole collisions likely account for the lower mobility.

The scattering of electrons between valleys of the conduction band in semiconductors plays an important role in determining the conductivity of the semiconductor, and has important implications for the physics of many high-speed devices. In the polar semiconductors GaAs and InP, the effective mass of the zone center ($\Gamma$) valley is lower than that of satellite valleys in the (111) and (100) directions ($L$ and $X$, respectively), producing high mobility for $\Gamma$ electrons and quite low mobility for conduction electrons in the satellite valleys. In the presence of high dc electric fields, the difference in mobility gives rise to the well-known Gunn effect. Recently, intervalley scattering in polar semiconductors has been studied on subpicosecond time scales using time-resolved luminescence spectroscopy,[1] differential transient absorption spectroscopy,[2] and electro-optic sampling of the reflection of a conical Cherenkov infrared shock wave.[3] These experiments indicate that a large fraction of electrons injected in the $\Gamma$ valley by femtosecond pulses near 620 nm scatter out of the $\Gamma$ valley within 100 fs, but require a few picoseconds to return. Consequently, the conductivity of photoexcited material increases as electrons return to the $\Gamma$ valley, achieving a steady high value until electron-hole recombination reduces the carrier density.

In this letter, we present new results on the mobility of photoexcited electrons in GaAs and InP probed in transmission by 200-fs infrared pulses in the range 15–100 $cm^{-1}$(50 $cm^{-1}$ = 1.5 THz = 6.2 meV). For injected electron densities in the range $3\times10^{16}$–$4\times10^{18}$ $cm^{-3}$, the transmission of the infrared probe pulse shows a rapid, pulse-limited decrease consistent with an insulator-to-metal transition in the photoexcited layer near the surface. The fast drop is followed by a more gradual ($\sim$1–2 ps) further decrease in transmission. The relative magnitude of this slow component is observed to increase with the photon energy of the excitation pulse, indicating more efficient scattering to satellite valleys at higher excitation energy. Furthermore, as the excitation energy increases to permit scattering to the $X$ valleys in GaAs, the time constant of the slow component of the transmission drop increases, suggesting that the effective return time of electrons to the $\Gamma$ valley is governed by the energy loss rate of conduction electrons.

The samples used in these experiments were 0.5-mm-thick semi-insulating wafers of GaAs and InP with (100) orientation. The infrared probe pulses were generated by reflecting 100-fs pulses ($\lambda$ = 630 nm, $E\approx200$ $\mu$J) from an InP (111) crystal near Brewster's angle.[4] The probe pulses were focused with an off-axis parabolic reflector and incident at 35° on the back side of the sample. The transmitted infrared light was collected with a second parabolic reflector and detected with a bolometer held at 4.2 K.[4] Excitation pulses of 100-fs duration were obtained by continuum generation followed by a 10-nm bandpass filter to select the wavelength. The excitation beam was incident at the same 35° angle on the front face of the sample so that the reflected pump light and the transmitted infrared light were collinear. In this way, the time resolution of the experiment was limited only by the duration of the infrared pulse. Transmission spectra of the infrared probe were obtained by taking the Fourier transform of the first-order autocorrelation of the transmitted light.[4]

Figure 1 shows the temporal response of the infrared transmission, $T(t)$, following optical excitation at $\lambda$ = 660 nm producing electron densities of 0.4, 0.9, and $2\times10^{18}$ $cm^{-3}$, listed top to bottom. Here, the transmission is defined as the ratio of the detected infrared light in the presence of the excitation pulse to that detected with the excitation beam blocked. The time traces all show an initial drop following excitation having approximately the 200-fs duration of the probe pulse. After this rapid initial drop, the transmission continues to fall for a period of a few picoseconds, ultimately arriving at a level that persists for over 100 ps. For longer times, the response begins to recover as electrons and holes recombine. The magnitude of the total transmission change (for $t>7$ ps) is seen to increase with the density of injected carriers. However, over the excitation density range measured, the kinetics are essentially independent of density.

Since the electron effective mass is much lower in the $\Gamma$ valley than in the $L$ and $X$ valleys, the drop in transmission and the associated rise in conductivity and mobility are due primarily to the density of electrons in the $\Gamma$ valley.[3] Fig-

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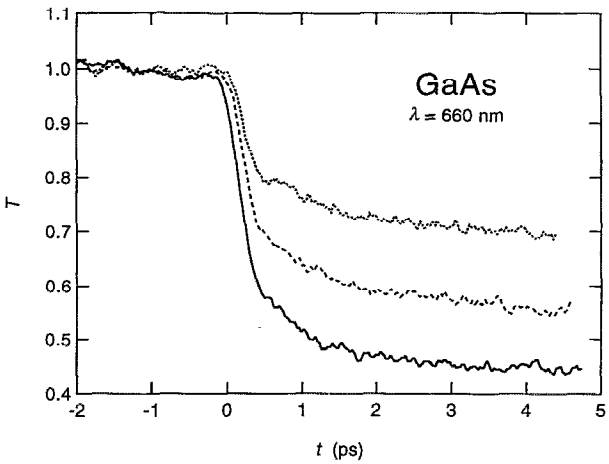


FIG. 1. The transmission $T$ of a 200-fs infrared probe pulse as a function of probe delay for GaAs excited with pump pulses at 660 nm. The photoexcited carrier densities near the surface are 0.4, 0.9, and $2\times10^{18}$ cm$^{-3}$ for the three traces, taken from top to bottom.

ure 2 shows the influence of the excitation photon energy on the transmission and Γ-electron kinetics. In the inset, the infrared transmission is plotted as a function of time for excitation at 580 nm ($N=6\times10^{17}$ cm$^{-3}$), 700 nm ($5\times10^{17}$ cm$^{-3}$), and 660 nm ($9\times10^{17}$ cm$^{-3}$), listed top to bottom. The figure clearly shows that the relative importance of the fast and slow components of the mobility rise depends on the excitation wavelength. This is consistent with increased scattering of electrons out of the Γ valley at higher excitation energy. At 700 nm, electrons are injected only up to the bottom of the $L$ valleys, whereas at 580 nm electrons can scatter to both $L$ and $X$ valleys. From the density dependence of the transmission the fraction of electrons in the high-mobility Γ valley can be calculated. This is shown in Fig. 2.

At an excitation wavelength of 700 nm, approximately 60% of the electrons remain in the Γ valley following excitation. The remainder return from the $L$ valleys with an

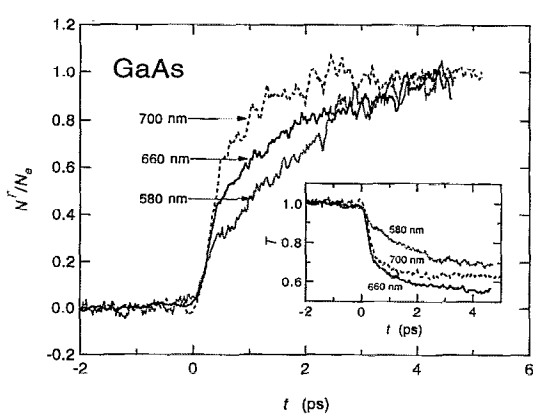


FIG. 2. Density of electrons in the Γ valley as a function of probe delay for excitation at 700 (broken line), 660 (solid line), and 580 nm (dotted line). The photoexcited carrier densities near the surface are 5, 9, and $6\times10^{17}$ cm$^{-3}$, respectively. The inset shows the measured transmission curves $T(t)$ from which the density traces were calculated.

exponential time constant of 0.7 ps. The $L$–Γ scattering rate is significantly lower than the Γ–$L$ scattering rate due to the higher density of final states in the $L$ valleys. As the wavelength is decreased, a greater fraction of electrons scatters out of the Γ valley within the duration of the probe pulse. At 580 nm, only about 25% of the electrons remain in the Γ valley. This is in general agreement with Monte Carlo simulations, which show more than 80% of the injected electrons scatter out of the Γ valley within 250 fs for excitation at 588 nm.[5] Furthermore, the time constant for the return of electrons to the Γ valley increases to 2 ps.

The longer return time can be explained by electron cooling rates in the following way. Electrons above the $L$-valley minimum can scatter back to the Γ valley by absorbing or emitting a high-momentum LO phonon. But the probability that they will quickly scatter back to the $L$ valley is high, driven by the much greater density of states in the $L$ valleys as indicated above. An electron at the bottom of the $L$ valley that scatters to the Γ valley by emitting a phonon, however, can return to the $L$ valley only by absorbing a phonon. Hence, only after the $L$ electrons have cooled to the bottom of the valley so that Γ–$L$ back transfer is reduced by degeneracy and the loss of phonon emission as a scattering channel, will there be appreciable $L$–Γ transfer. The cooling rate due to LO phonon emission can be estimated from the LO phonon energy, $\epsilon_{\rm ph}=35$ meV, and emission time, $\tau_{\rm ph}\approx0.17$ ps.[6] The cooling time for electrons injected at energy $\epsilon$ above the Γ minimum is then $t=\tau_{\rm ph}(\epsilon-\epsilon_{\Gamma L}/\epsilon_{\rm ph})$. This gives a time of 0.6 ps for electrons excited from the heavy-hole band with 660-nm pulses and 1.7 ps for 580-nm pulses, in good agreement with the observed rates.

To obtain a quantitative estimate of the mobility of the photoinjected electrons, the infrared transmission spectra taken 7 ps after excitation were compared to spectra of unexcited material. Due to a stray reflection inside the bolometer, the spectra display an oscillatory artifact. The spectrum of probe light is shown at the bottom of Fig. 3 and extends from roughly 15 to 100 cm$^{-1}$, centered at $\sim$40 cm$^{-1}$. For the range of carrier densities from 0.4 to $5.3\times10^{18}$ shown in Fig. 3, the transmission depends only weakly on frequency.

To extract the electron mobility from the infrared transmission, the spectra were fitted to the transmission calculated for a conducting plasma assuming a Drude model for the conductivity. This simple model has been applied very successfully to similar far-infrared (FIR) transmission experiments on uniformly doped silicon samples.[7] According to the Drude model, the complex dielectric constant $\epsilon=\epsilon_r+i\epsilon_i$ is given by the expression $\epsilon=\epsilon_0+(4\pi\sigma i)/\omega$, where $\epsilon_0$ represents the contribution of the bound charges and the frequency-dependent conductivity $\sigma(\omega)$ is given by $\sigma(\omega)=(Ne^2)/[m(\Gamma-i\omega)]=(\sigma_{dc})/(1-i\omega/\Gamma)$. Gaussian units are used in these expressions, $m$ is the effective mass of the electrons, $\omega$ is the angular frequency of the light, Γ is the scattering rate, and $\sigma_{dc}$ is the conductivity at zero frequency. Because of their higher mass and stronger coupling to LO phonons, the holes contribute little to the con-

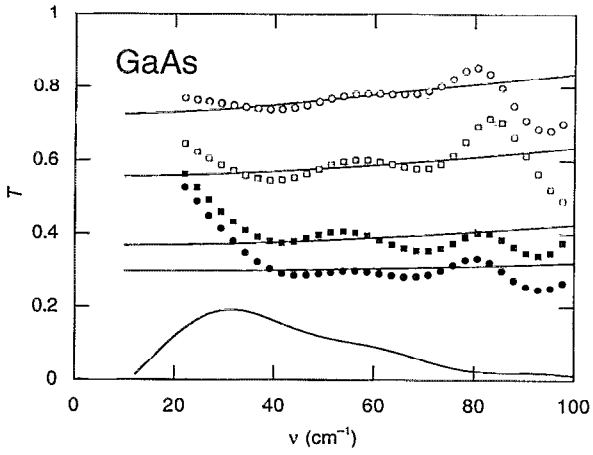


FIG. 3. Infrared transmission spectra of photoexcited GaAs taken 7 ps after excitation with 660-nm pump pulses. From top to bottom, the injected electron densities are 0.4, 1.2, 2.8, and $5.3\times10^{18}$ cm$^{-3}$. The spectrum of the infrared probe pulse in the absence of excitation is shown at the bottom. Lines through the data points are single-parameter fits to a Drude model giving electron mobilities of 1300, 870, 720, and 480 cm$^2$/Vs.

ductivity and this contribution has been ignored. The electron mobility is then given by $\mu = \sigma_{dc}/eN = e/m\Gamma$.

The photoexcited plasma layer is thin compared to the probe wavelength so that the transmission can be calculated from the thin film equations.[8] The Drude fits are shown in Fig. 3 and yield scattering rates $\Gamma/2\pi$ of 100, 160, 190, and 290 cm$^{-1}$ for electron densities of 0.4, 1.2, 2.8, and $5.3\times10^{18}$ cm$^{-3}$, respectively. These correspond to electron mobilities of 1300, 870, 720, and 480 cm$^2$/Vs, which are 2.5–4 times lower than the ~2000–3000 cm$^2$/Vs values for $n$-doped GaAs at room temperature and these densities.[9] These values are significantly lower than those reported by Nuss *et al.*[3] but are similar to some recent results.[10] The results of similar experiments on InP were qualitatively similar, with photoinjected electron mobilities 2–3 times lower than equilibrium $n$-doped values.

The photoexcited carrier plasma differs from the $n$-doped case in two important respects. First, the dominant carrier cooling mechanism is by the emission of low-momentum LO phonons. A nonequilibrium distribution of LO phonons is established several picoseconds after excitation at these densities as electrons and holes relax towards the band edge.[11] The hot phonon distribution increases electron-phonon scattering, reducing the mobility. This effect is unlikely to be large at room temperature, however, because the relative change in the LO phonon population is small. Second, photoexcitation produces an equal density of electrons and holes. Since the hole mass is much greater than the electron mass ($m_e = 0.067\ m_0$, $m_{hh} = 0.57\ m_0$), electron-hole scattering is inefficient in transferring energy from the electrons to the holes, but effective in randomizing electron momentum, thereby decreasing the electron mobility.[12] The importance of electron-hole scattering increases with density, leading to the factor of 4 decrease observed at the highest density investigated.

In conclusion, we have used 200-fs pulses of far-infrared radiation to study the mobility of photoexcited electrons in GaAs and InP. For excitation with visible light in the wavelength range 560–700 nm, the transmission of infrared probe light undergoes a rapid (~200-fs) decrease following excitation, followed by a slower (0.7–2 ps) rise to a steady value that persists longer than 100 ps. The drop in transmission is explained by the increasing conductivity of optically injected free-carrier plasma near the surface as electrons settle in the high-mobility $\Gamma$ valley. Photoexcitation at energies above the $X$ valley minima in GaAs leads to increased scattering out of the $\Gamma$ valley and longer return times. Infrared transmission spectra taken 7 ps after excitation show weak frequency dependence over the range 25–80 cm$^{-1}$ and have been analyzed in the context of a thin film of Drude plasma at the surface. The model takes into account the effect of the plasma on both real and imaginary parts of the dielectric constant, as well as the finite thickness of the excited region. The fits yield values of mobility that are 2–4 times lower than those observed in $n$-doped samples at comparable densities.

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