

Anomalous reduction in the energy loss of electrons in *p*-type semiconductors

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We report the observation of an anomalous decrease in the energy-loss rate of minority electrons in *p*-type GaAs at doping levels above $p > 2 \times 10^{19} \text{ cm}^{-3}$. This unusual behavior of the electron-hole energy transfer is a general property of plasmas at high carrier densities and is due to the transition from dominant plasmon-mediated scattering to free-carrier-screened single-particle scattering. This effect is observable when the excess energy of hot carriers interacting with a cold plasma is comparable to the plasmon energy $\hbar\omega_{\text{pl}}$. [S0163-1829(97)52612-9]

The energy relaxation of minority carriers in semiconductors has recently been studied in a series of experiments in modulation-doped quantum wells^{1,2} and in doped GaAs.³⁻⁸ These previous experiments have shown that electron-hole (*e-h*) scattering is an important energy-loss channel for minority carriers. In a recent experiment on *p*-type GaAs,⁸ an extremely fast (< 200 fs) onset of the band-edge luminescence due to ultrafast cooling of minority electrons within the central conduction-band valley has been reported. From this finding and time-resolved spectra it could be shown that at high doping levels ($\sim 10^{19} \text{ cm}^{-3}$) the energy-loss rate due to *e-h* scattering is considerably higher than the rate due to LO-phonon emission.⁸

In *p*-type GaAs at low and moderate doping levels it has also been shown that the energy transfer by *e-h* scattering increases monotonically with increasing doping concentration.^{3,5} In the high doping regime, to our knowledge, the dependence of the energy loss on doping concentration has not been investigated until now. The behavior of the electron mobility and momentum relaxation is known from transport measurements⁹⁻¹¹ and theoretical calculations.^{12,13} However, no direct observation of the femtosecond energy relaxation of minority electrons has been reported for doping concentrations above $p > 2 \times 10^{19} \text{ cm}^{-3}$.

The effect of a high concentration of majority holes on the energy relaxation of minority electrons raises a very interesting fundamental question. How does the increase in the number of holes and subsequent increase of hole degeneracy, hole plasma frequency, and screening influence the electron energy-loss rate? In particular, how does the electron energy loss behave when the electron energy is comparable to the hole plasma energy, $\hbar\omega_{\text{pl}}$? To our knowledge, the energy transfer in this parameter range has never been experimentally investigated in a plasma.

This paper presents experimental evidence for a *reduction* in the energy relaxation rate of minority electrons in *p*-type GaAs at very high doping densities. We have performed time-resolved luminescence measurements on *p*-type GaAs samples with different doping concentrations. The experimentally determined rise time of the band-edge lumines-

cence exhibits a minimum at a hole density of $p \sim 2 \times 10^{19} \text{ cm}^{-3}$, indicating that the energy loss of minority electrons is reduced at very high doping levels. Monte Carlo simulations of our experiments reveal that this minimum occurs for doping densities at which the characteristics of the *e-h* scattering change from plasmon mediated to quasistatically screened, resulting in a nonmonotonic behavior of the energy-loss rate.

As an experimental tool to monitor the energy loss of photoexcited minority electrons, we use the temporal evolution of the luminescence spectra. The extremely fast dephasing process of free carriers in the case of dominant carrier-carrier scattering [phase relaxation time ~ 10 fs (Ref. 14)] allows a direct description of the luminescence spectra through carrier distribution functions. The luminescence intensity corresponding to the optical transition energy $h\nu$ at time t is given by a sum over the heavy- and light-hole contributions:¹⁶

$$I_{\text{lum}}(h\nu, t) \propto (h\nu)^2 \sum_{h=\text{hh, lh}} |H_{\text{opt}}^{(h)}|^2 \rho(h\nu) \times f_e(E_e^{(h)}, t) f_h(E_h^{(h)}, t), \quad (1)$$

where $f_e(E_e^{(h)}, t)$ and $f_h(E_h^{(h)}, t)$ are the electron and hole distribution functions at the corresponding transition energies, $|H_{\text{opt}}^{(h)}|^2$ is the transition matrix element, and $\rho(h\nu)$ is the joint density of states. Since in our experiments we work with low excitation densities and high doping concentrations, the hole distribution function remains practically at equilibrium and therefore constant over time. As a consequence, the temporal evolution of the luminescence is exclusively determined by the time evolution of the electron distribution function.

In the experiments we used a series of six different *p*-type GaAs samples with doping concentrations of $1.0 \times 10^{18} \text{ cm}^{-3}$ (sample A), $1.0 \times 10^{19} \text{ cm}^{-3}$ (sample B), $1.8 \times 10^{19} \text{ cm}^{-3}$ (sample C), $1.9 \times 10^{19} \text{ cm}^{-3}$ (sample D), $2.7 \times 10^{19} \text{ cm}^{-3}$ (sample E), and $4.0 \times 10^{19} \text{ cm}^{-3}$ (sample F). All the samples were grown by molecular-beam epitaxy on a semi-insulating (100) GaAs substrate; the GaAs layers are C or Be doped. The samples A, B, D, and F are

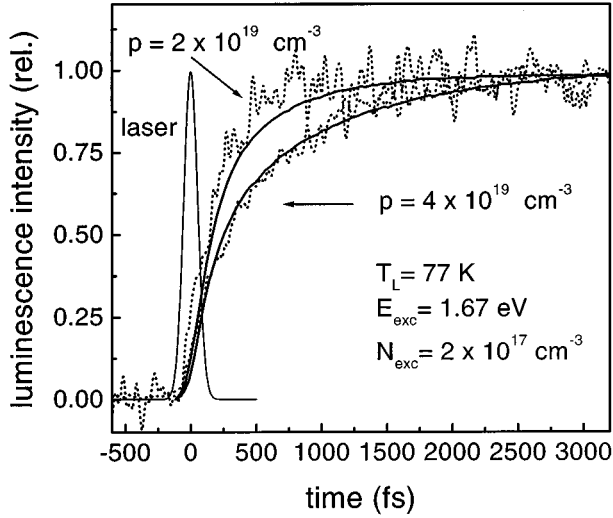


FIG. 1. Time evolution of the luminescence intensity at 13 meV above the renormalized band edge for sample *D* ($p=1.9\times 10^{19}\text{ cm}^{-3}$) and sample *F* ($p=4.0\times 10^{19}\text{ cm}^{-3}$). The solid lines show the results of the ensemble-Monte Carlo simulations.

$p\text{-Al}_x\text{Ga}_{1-x}\text{As}/p\text{-Al}_x\text{Ga}_{1-x}\text{As}$ double heterostructures with a mole fraction of $x=0.5$. The thickness of the doped GaAs region was 1000 nm for samples *B*, *E*, and *F*; 500 nm for samples *A* and *C*; and 125 nm for the sample *D*.

Our time-resolved luminescence experiments were performed using a Kerr-lens mode-locked Ti-sapphire laser at a repetition rate of 76 MHz. The pulse duration was approximately 80 fs and the peak of the excitation wavelength was varied between 738 and 800 nm to keep the electron excitation energy below the GaAs satellite valleys and the total excitation energy below the band gap of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ cladding layers. The samples were mounted in a liquid-nitrogen-cooled cryostat ($T_L=77\text{ K}$). In order to obtain a time resolution below 100 fs, we used the upconversion technique¹⁵ with a dispersionless parabolic mirror system to collect and focus the sample luminescence. The luminescence signal was upconverted using a 1-mm beta barium borate (BBO) crystal and type I phase matching. The sum-frequency signal was dispersed by a monochromator and detected by a cooled GaAs photomultiplier coupled to a photon counting unit. The experiments were performed with very low injected carrier densities ($N_{\text{exc}}\approx 1\times 10^{17}\text{ cm}^{-3}$) as compared to the doping background.

We first measured the time evolution of the luminescence intensity at various photon energies for the different samples. Figure 1 shows the onset of the experimental luminescence intensity at the luminescence maximum, i.e., $\approx 13\text{ meV}$ above the band edge, for sample *D* with $p=1.9\times 10^{19}\text{ cm}^{-3}$ and for sample *F* with $p=4.0\times 10^{19}\text{ cm}^{-3}$. The rise time for sample *F*, our highest doped sample, is clearly *slower* than that for the sample with the lower doping concentration. Figure 2 shows the experimental band-edge luminescence rise time (left axis) versus doping concentration, as obtained for our six samples. Here the rise time is defined as the time it takes for the luminescence intensity to reach 75% of its maximum value, with the time zero taken at the

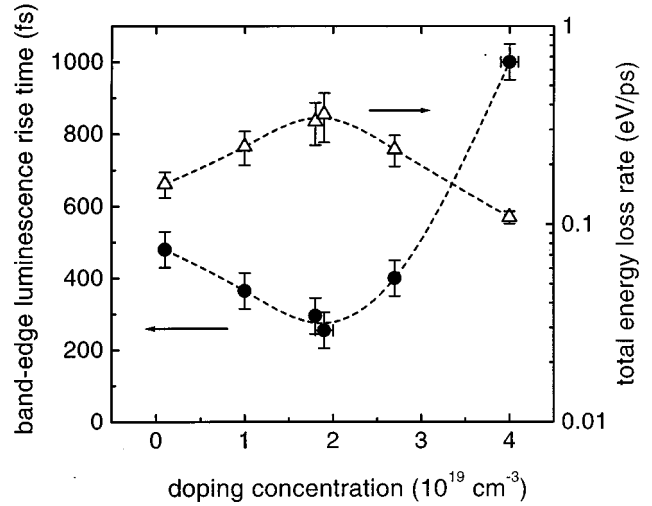


FIG. 2. Band-edge luminescence rise time (left axis) and corresponding average of the total energy loss rate of electrons as defined in the text (right axis) vs doping concentration. The dashed lines are only to guide the eye.

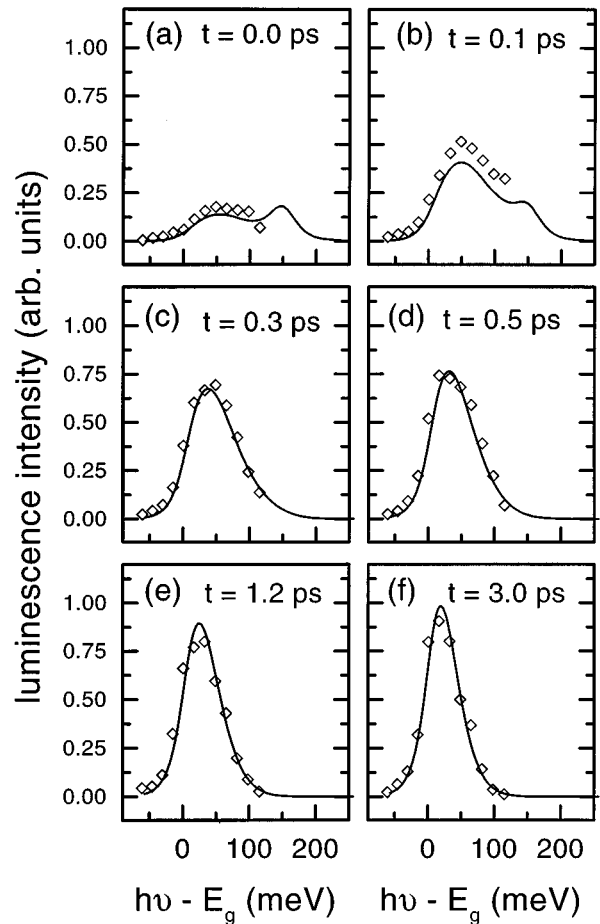


FIG. 3. Luminescence spectra at a delay time of (a) 0.0 ps, (b) 0.1 ps, (c) 0.3 ps, (d) 0.5 ps, (e) 1.2 ps, (f) 3.0 ps for sample *F* ($p=4.0\times 10^{19}\text{ cm}^{-3}$). The solid lines are the results of our ensemble-Monte Carlo simulations.

center of the excitation pulse. A clear minimum is observed for the doping concentration of $p \approx 2.0 \times 10^{19} \text{ cm}^{-3}$.

In order to determine the relationship between the electronic energy relaxation and the rise time of the band-edge luminescence in more detail, we analyzed the luminescence spectra at different delay times. The symbols in Fig. 3 show the measured luminescence spectra for sample *F* along with the theoretical Monte Carlo results (solid lines) at six characteristic delay times. Nonthermal features are not present in the observed spectra. Thus, thermalization occurs on the time scale of our experimental resolution (< 100 fs).

This observed extremely fast thermalization is in agreement with earlier measurements¹⁷ at similar excitation densities and is also supported by our theoretical analysis discussed below. In order to obtain a quantitative measure of the electron energy relaxation, we first interpret the band-edge luminescence data (Figs. 1 and 2), assuming a fully thermalized electron distribution function. Since effects of electron degeneracy are of minor importance at these low excitation densities, f_e can be well approximated by an internally thermalized Maxwell distribution with a transient electron temperature $T_e(t)$ and a corresponding mean electron energy $\langle E_e(t) \rangle = 3k_B T_e(t)/2$, where k_B is Boltzmann's constant. From Eq. (1) we see that the band-edge luminescence intensity is an extremely sensitive measure of the transient electron mean energy.

In order to determine the energy-loss rate as a function of the doping density, we analyzed the time interval in which the band-edge luminescence increases to 75% of its maximum value. By use of a heated Maxwellian f_e in Eq. (1) and an appropriate frequency convolution to account for the spectral laser width and the experimental spectral resolution of 35 meV, one obtains $T_e(t)$ as a function of the measured band-edge luminescence intensity. Thus, 75% of the maximum band-gap luminescence corresponds to an average electron energy of approximately 17 meV ($T_e = 130$ K) for all investigated densities. The resulting total energy-loss rate of electrons is given by the difference between their initial excess energy of ≈ 117 meV and their average energy at $T_e = 130$ K, divided by the luminescence rise time. The obtained energy-loss rate as a function of doping concentration is shown in Fig. 2 (right axis). The energy loss exhibits a maximum at the majority hole concentration of $p = 1.9 \times 10^{19} \text{ cm}^{-3}$. At higher doping concentrations, the total energy-loss rate of electrons dramatically decreases.

It is well known from related earlier work that the electron-hole interaction is the dominant scattering mechanism in the energy loss of minority electrons at such high doping levels.^{3–8} One expects that the electron-hole scattering rate would increase with increasing doping and hence result in an increase in the number of scattering partners. However, our observation of a decrease in the total energy-loss rate suggests that the electron-hole scattering becomes less effective as an energy loss channel at higher doping levels, resulting in a decreased total energy-loss rate of minority electrons and a slower rise of the band-edge luminescence.

In order to determine the contributing effects responsible for the decrease in the electron-hole scattering, the dynamics of the carriers was quantitatively analyzed using ensemble–Monte Carlo simulations (solid lines in Figs. 1 and 3).

The underlying transport model had to account for the following specific features of our experiments: (i) the electron excitation energies were of the order of the plasmon energies $\hbar \omega_{\text{pl}}$ of the doping background, calling for a fully dynamical free-carrier screening of all long-range carrier interactions; (ii) because $\hbar \omega_{\text{pl}}$ was of the order of the average energy separation between the heavy-hole (hh) and light-hole (lh) valence band, we had to go beyond the simple parabolic valence-band model to provide an accurate description of plasmon-induced $\text{hh} \leftrightarrow \text{lh}$ transfers in upper band regions; (iii) due to the high doping levels, hole degeneracy could be expected to play a decisive role in the electron-hole dynamics; and (iv) in accordance with earlier work,¹⁸ impurity banding strongly dominates over any bound hole contributions (with the possible exception of the moderately doped sample *A*). This is experimentally confirmed by Hall data for samples *D* and *F* which show a negligible temperature dependence between 10 and 300 K.

Since the technical details of our ensemble–Monte Carlo approach have been described in a previous paper,⁷ we only note the novel implementation of a more detailed valence-band structure (treated within an $8 \times 8 \vec{k} \cdot \vec{p}$ model¹⁹) and of a full dynamical dielectric function $\epsilon_{\text{RPA}}(q, \omega)$ using the random phase approximation (RPA).²⁰ Given the spectral laser width and the experimental spectral resolution, a corresponding frequency convolution ($\Delta h\nu \approx 35$ meV) of the calculated luminescence spectra was performed.

Let us first discuss our findings regarding the transient luminescence. Quite generally one observes two relaxation regimes: (i) an initial extremely fast thermalization and an energy dissipation stage at times below typically 200 fs dominated by very efficient plasmon-mediated e - h scattering, and (ii) an ensuing much slower cooling dynamics governed by the “conventional” (i.e., quasistatically screened) single-particle collisions, with a pronounced retardation due to strong hole degeneracies.

The initial stage with its high energy dissipation rates $\langle dE/dt \rangle$ up to ~ 0.8 eV/ps is due to very pronounced inter-valence-band contributions to both the direct scattering dynamics and the dielectric response.^{20,21} Typically 80% of $\langle dE/dt \rangle$ occurs via the $\text{hh} \rightarrow \text{lh}$ scattering channel, whereas the $\text{hh} \leftrightarrow \text{lh}$ contributions to $\epsilon_{\text{RPA}}(q, \omega)$ lead to a strong broadening of the plasma resonance. The direct comparison with our experimental data in Figs. 1 and 3 shows excellent agreement for this initial regime. In particular, our theoretical findings confirm that the electron distributions are fully thermalized after 200 fs and lead to the fast onset of the band-edge luminescence.

We now turn to the physical interpretation of the reduction in the electron energy relaxation rate at doping densities $p > 2 \times 10^{19} \text{ cm}^{-3}$. We can distinguish two different tendencies depending on the mean energy $\langle E \rangle$ of the electron distribution. For $\langle E \rangle$ well above the plasmon emission threshold, $\langle dE/dt \rangle$ increases with p due to the increasing plasmon energy and plasmon emission rate, while for $\langle E \rangle$ far below $\hbar \omega_{\text{pl}}$, an opposite trend prevails because of the strong quasistatic screening of the e - h interaction. The transition region between these two asymptotic tendencies displays a maximum of $\langle dE/dt \rangle$ as a function of p , which in our simulations turns out to be roughly at $\hbar \omega_{\text{pl}}(p) \approx \langle E \rangle / 2$. We emphasize that a similar behavior can be found in the theory of the

free-electron gas,²² which demonstrates that this is a general property of free-carrier plasmas.

Our simulations well reproduce the temporal evolution of the luminescence intensity for photon energies above and around the maximum of the luminescence spectra for all doping densities (see Fig. 1). At the band edge, the calculated luminescence intensity shows a slower rise as compared to the measured intensity for all samples. We attribute this systematic discrepancy to (i) our neglect of the interplay of scatterings with the coherent excitation dynamics,^{23,24} which should lead to faster initial thermalization of the electrons, (ii) our treatment of the (quasistatistically screened) electron-electron interactions, and (iii) possible effects of impurity banding. Nevertheless in accordance with our experimental findings, all simulations revealed a pronounced increase of the luminescence rise time at doping levels above $p=2 \times 10^{19} \text{ cm}^{-3}$.

We note that a minimum in the mobility of electrons in *p*-type GaAs at a doping concentration of $p=1 \times 10^{19} \text{ cm}^{-3}$ has been reported in the literature¹⁰ and was explained by a reduction of the *e-h* scattering at higher doping concentrations.¹³ Thus, momentum relaxation and energy loss of minority electrons at very high hole densities reveal consistent behavior.

In conclusion, we have observed a decrease in the energy-loss rate of minority electrons in *p*-type GaAs for doping concentrations above $p=2 \times 10^{19} \text{ cm}^{-3}$. This nonmonotonic behavior of the energy-loss rate as a function of doping concentration is due to the transition from plasmon mediated *e-h* scattering at lower densities to quasistatic screening of the *e-h* interaction by the degenerate hole plasma at high densities. This effect is observable when the excess energy of hot carriers interacting with a cold plasma is comparable to the plasmon energy $\hbar \omega_{\text{pl}}$ and is important for understanding systems of high plasma density. To our knowledge the present analysis constitutes the first direct experimental evidence for the density dependence of the energy-loss rate of energetic particles in an equilibrium background plasma of very high density and low temperature.

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