RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 8 15 AUGUST 1993-II

Femtosecond hole relaxation in *n*-type modulation-doped quantum wells

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(Received 7 June 1993)

We present a study of femtosecond hole-relaxation dynamics in *n*-modulation-doped $GaAs/Al_xGa_{1-x}As$ quantum wells at low temperatures and low photoexcitation density. We conclude that holes are nonthermal for approximately the first 800 fs and determine the hole-electron energy loss rates by comparing experimental results with Monte Carlo simulations. These results represent a definitive study of hole scattering and relaxation processes in a semiconductor.

Femtosecond spectroscopy provides a powerful technique for investigating ultrafast relaxation of photoexcited carriers in semiconductors.[1-6] In most studies, however, the optical response of the system is determined primarily by electrons and very little is known about hole-relaxation dynamics, except for some recent results on bulk GaAs at room temperature.[7] Earlier measurements[1] of differential transmission spectra of modulation-doped quantum wells at room temperature also provided information primarily about electron relaxation processes.

The key to study the hole dynamics is to devise an experiment in which the hole contribution dominates the optical response. The basic idea is to investigate *n*-modulation-doped quantum wells at low temperatures, low excitation densities, and low excess photon energy so that the photoexcitation does not significantly perturb the well-defined Fermi distribution of electrons at low electron energy.[8] Therefore, the luminescence dynamics is dominated by the hole dynamics. Furthermore, optical-phonon scattering is suppressed in the highest valence band so that the dynamics is dominated by hole-hole and hole-electron scattering so that unambiguous information about these processes can be obtained from such measurements.

We conclude from our measurements that photoexcited holes remain in a nonthermal distribution for approximately 800 fs following the optical excitation. We present Monte Carlo simulations that provide good agreement with the experiments. A comparison between the experiments and the simulation allows us to conclude that hole-electron, and to a lesser extent hole-hole, interactions play the dominant role in hole relaxation and to quantitatively determine the electron-hole energy exchange rate.

The experiments are based on femtosecond luminescence up-conversion.[9] The sample consists of 60 pairs of 5-nm-thick GaAs wells and $Al_{0.3}Ga_{0.7}As$ barriers. Part of the barrier layers was doped with Si and provided 5×10^{11} cm^{-2} electron concentration in the quantum wells (corresponding to a Fermi energy of about 18 meV). The sample, mounted on a 5-K cold finger of a cryostat, was excited by pulses (duration <200 fs) from a mode-locked Ti:sapphire laser operating at 1.78 eV. The luminescence and the second laser pulses were focused onto 4-mm-thick $LiIO_3$ to generate the sum frequency. The up-converted signals were analyzed in a double monochrometer (resolution 8 meV), and detected by single-photon counting. The time resolution is determined mainly by the pulse duration, and is approximately 200 fs. The excitation density was 1×10^{10} cm^{-2}, estimated from the absorption coefficient of $GaAs/Al_xGa_{1-x}As$ quantum well at 1.78 eV and the spot size of the laser beam. The accuracy of the excitation density is approximately a factor of 2.

In order to obtain quantitative information from the experiments, we calculated the dispersion of the valence bands using the Luttinger-Kohn Hamiltonian under the axial approximation.[10] Using these nonparabolic dispersion curves (see the inset of Fig. 1), we calculate that the laser pulses at 1.78 eV create electrons at 140 meV from the band edge by electron–heavy-hole (HH) transition (band gap 1.619 eV), and at 123 meV by electron–light-hole (LH) transition (band gap 1.64 eV, corresponding to HH-LH valence-band separation of 21 meV at $k=0$). Heavy and light holes are created with excess kinetic energy of 21 and 17 meV, respectively.

The upper limit of our estimate of photoexcited density is 2×10^{10} cm^{-2}, so that there are at least 25 electrons per photoexcited electron. Therefore, the maximum excess energy given to the electron system is 140/25 or 5.6 meV per electron. A simple calculation, as well as Monte Carlo simulations, show that this corresponds to an electron temperature rise to at most 90 K, and a change of less than 10% in the electron occupancy near the band edge. Furthermore, simulations show that luminescence spectra are insensitive to electron temperature in this range. We conclude that the temporal evolution of band-edge luminescence is insensitive to electron dynamics and is determined by the dynamics of hole relaxation. *Therefore, our measurements provide direct information about*

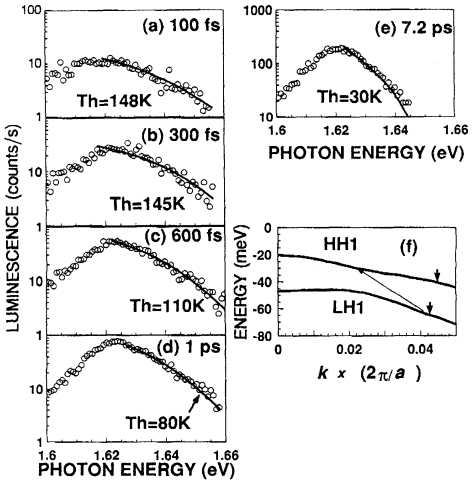

FIG. 1. Luminescence spectra of an n-type modulation doped $GaAs/Al_xGa_{1-x}As$ quantum well after femtosecond excitation at 1.78 eV. The luminescence intensity is plotted for delay time of (a) 100 fs, (b) 300 fs, (c) 600 fs, (d) 1 ps, (e) 7.2 ps. Circles represent experimental results. The lines show the best fits to the spectra assuming thermalized carriers with hole temperatures as shown and electron temperatures of 98, 87, 82, 80, and 30 K. The fits are not sensitive to electron temperatures in this photon energy range. (f) shows the calculated valence subband dispersion curves, with downward arrows indicating the initial position of photoexcited holes and the light arrow showing the LH-HH transition with an optical-phonon emission.

hole dynamics.

The excess kinetic energies of holes in the lowest subband are smaller than the optical-phonon energy[11] (33.3 meV for TO phonons and 36.2 meV for LO phonons), even if we consider the spectral width of the optical pulses. Intraband hole–optical-phonon scattering is therefore suppressed. However, light holes emit optical phonons and create a heavy-hole distribution at about 10 meV from the band edge. The dynamics of heavy holes is determined by interaction with cold electrons, other holes, and acoustic phonons.

We measured the luminescence spectra at several delay times and the temporal evolution of the luminescence at the band edge (1.62 eV). Figure 1 shows the luminescence spectra for five time delays ranging from 100 fs to 7 ps. Assuming **k** conservation, and an average effective HH mass from the **k·p** calculations, this spectral range corresponds to holes of energy <5 meV. Figure 2 shows the time evolution of the band-edge luminescence for two different time scales.

We first analyze the luminescence spectra assuming hot, thermalized distributions for the electrons (Fermi-Dirac) and holes (Maxwell-Boltzmann). Using the hole (electron) temperature T_h (T_e), the luminescence intensity[12,13] at photon energy $\hbar\omega$ is given by

$$I(\hbar\omega)=(A/k_BT_h)\exp[-(1-c)(\hbar\omega-E_g)/k_BT_h] \times(1/1+\exp[\{c(\hbar\omega-E_g)-E_F\}/k_BT_e]) , \quad (1)$$

where c is a parameter related to the electron–heavy-hole effective-mass ratio,

$$c=[1+m_e/m_h]^{-1} . \quad (2)$$

The electron dispersion was assumed to be parabolic with an effective mass of $0.067m_0$. The HH effective mass was taken to be $0.49m_0$, since the calculated valence-band dispersion curves can be fit to parabolas with the HH effective mass of $0.49m_0$ and a LH effective mass of $0.45m_0$, with an error of less than 3 meV over the entire region of interest. The fits to the spectra are not sensitive to the hole mass, implying that the nonparabolicity of the bands is also not important. We determined E_g, the band-gap energy for the first electron–heavy-hole subband transition, and the electron density from the luminescence spectrum at 14 ps, i.e., after electrons and holes reached quasiequilibrium. The values agreed well with the design values. We treated the quasi-Fermi level for electrons E_F as a function of electron temperature, though it did not affect the fitting. The parameter A is given by

$$A=\eta N_h/\rho_h , \quad (3)$$

where N_h is the hole density, ρ_h the hole density of states, and η a constant related to emission and detection efficiency.

The solid curves in Fig. 1 show the fit to the spectra obtained by assuming quasiequilibrium distributions [Eq. (1)]. Although both electron and hole temperatures were treated as fitting parameters, the hole temperature played the dominant role in the fitting near the band edge ($E-E_g<20$ meV) and the electron temperature only affected the tail at higher energies. This result is expected for a heated Fermi distribution, as discussed above. The fitting indicates that the hole temperature rose to 148 K at 100 fs and cooled down to 80 K at 1 ps. This thermalized hot-carrier model provides a reasonable fit to the spectra as shown in Fig. 1. However, using the calcu-

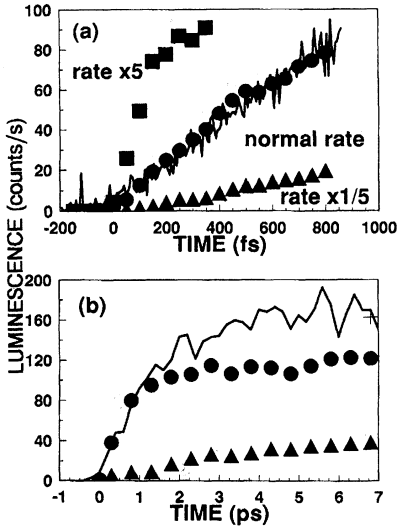

FIG. 2. Femtosecond (a) and picosecond (b) time evolution of the band-edge luminescence intensity. Solid curves show the experimental results. Circles represent the Monte Carlo simulation results using normal carrier-carrier scattering rate. Results using five times larger (squares) and five times smaller (triangles) scattering rates are also shown in (a). The cross in (b) shows the calculated intensity if the temperature is 30 K instead of 40 K at 7 ps. The triangles in (b) show the effect of turning off the electron-hole scattering.

lated valence-band structure, it is easy to show that the measured spectral range of 35 meV corresponds to holes in the *top* 5 meV of the HH valence band. Therefore, we may not conclude that the entire hole distribution is *thermal.* In fact, the temporal evolution of the luminescence intensity cannot be explained by a thermalized hole distribution, as we show below.

For a thermal hole distribution, A in Eq. (1) is a constant (so long as hole recombination can be neglected), and the band-edge luminescence intensity should vary inversely with the hole temperature [Eq. (1)]. Figure 2 shows the variation of the peak intensity of the near-band-edge luminescence as a function of time delay. From our known spectral resolution and the calculated dispersion curves, we calculate that only holes within 1 meV of the top of the HH1 valence band contribute to this intensity. We see that the intensity increases by about a factor of 7 between 100 and 1000 fs. However, the best fit to the spectral shape assuming a thermal model (Fig. 1) shows that the hole temperature decreased by less than a factor of 2 (from 148 to 80 K) in this delay range. Furthermore, the maximum initial hole temperature for our photoexcitation condition is <300 K so that we would need a temperature of 40 K or less at 1000 fs to explain the variation of luminescence intensity between 100 and 1000 fs. The spectral shapes simply cannot be fit to a temperature of 300 K at 100 fs and 40 K at 1000 fs. Therefore, the thermalized hot-hole distribution model *cannot* explain the measured temporal variation of the luminescence intensity in the first 1000 fs. An approximate estimate of the fraction of nonthermal holes may be obtained by using A as an adjustable parameter in fitting the variation of the peak luminescence intensity with time delay. If we assume that all holes are thermalized at 1000 fs, then our fits show that only 33%, 67%, and 80% of the holes are thermalized at 100, 300, and 800 fs, respectively. We, therefore, conclude that a substantial ($\geq 20\%$) fraction of the holes are *nonthermal* for nearly 800 fs.

There is no inconsistency between the facts that the spectra can be fit to a thermalized hole distribution, and the fact that temporal variation of the near-band-edge intensity requires a *nonthermal* hole distribution for the first 1000 fs. Since the spectra correspond to only holes within about 5 meV of the top of the valence bands, the first fact implies that the distribution of holes *in this energy range* can be *approximated* by a thermal distribution within the uncertainties of the experiments and the fitting procedure. The spectra provide no information about holes at higher energies. It is the thermalization and relaxation of these high-energy holes that lead to the large increase in the band-edge luminescence intensity in the first 800 fs.[14]

Figure 2(b) shows that the band-edge luminescence intensity reaches half the maximum intensity at 1 ps, and then increases slowly and reaches a maximum at about 7 ps. Since optical-phonon emission is not important for the heavy holes, we attribute the rise in the first 1 ps to the hole thermalization and the energy loss of holes by electron-hole scattering. After 1 ps, electrons and holes are in quasiequilibrium with a common temperature, and the slow increase in intensity beyond 1 ps is due to the cooling of this thermalized electron-hole plasma by phonon emission.

In order to make these arguments more quantitative, we performed an ensemble Monte Carlo[15] (EMC) simulation of the dynamics of carriers using a three-band model, consisting of electrons, heavy holes, and light holes in their first subbands. The bands are treated as parabolic with in-plane effective masses as indicated above. We assume the absorption coefficient ratio to be 2:1 for HH-electron and LH-electron transition. In the present MC, we use bulk phonon modes to calculate the carrier–optical-phonon scattering. Hot-phonon effects are not included but degeneracy is included. The carrier-carrier scattering rate is calculated using a time-dependent static screening constant, determined by the instantaneous carrier distribution function, as discussed elsewhere.[13] Dynamic screening is shown to be important for electrons interacting with a thermalized electron-hole plasma.[16,17] However, we expect that the static screening approximation would be adequate because holes have large masses, and because we estimate that the average hole energy becomes smaller than the average electron energy for times >150 fs. Since no calculations of dynamic screening for the athermal case have been reported, we defer detailed consideration of dynamic screening effects to future work.

Figure 3 shows (as solid dots) the calculated HH distribution function (DF) as a function of hole energy at 100, 300, and 500 fs after the pulse peak. The solid curves show the Maxwellian DF with a temperature corresponding to the average hole energy. The hole DF is distinctly *nonthermal* at 100 fs. It shows two peaks, one due to photoexcited HH's, and the other resulting from the optical-phonon replica of photoexcited LH's. After 300 fs, the DF becomes much broader but still clearly nonthermal. Almost all the holes near the band edge are thermalized after 500 fs. The degree of thermalization at the band edge, which is estimated by the ratio of hole occupancy to the thermalized one, agrees well with the experiment.

The temporal evolution of the luminescence calculated by EMC agrees well with the experiment, as shown in Fig. 2(a) (circles). Figure 2(a) also shows the calculated time evolution with carrier-carrier scattering rate artificially increased by a factor of 5 (squares) and decreased five times (triangles) than the calculated rate. The time evolution depends sensitively on the carrier-carrier scattering rate, indicating that the time evolution is determined by the hole thermalization and the energy dissipation to the cold electrons. This point is further verified by artificially shutting off the electron-hole interaction which leads to much slower relaxation as shown in Fig. 2(b) (triangles). In contrast, we find that shutting off hole-hole scattering has little effect on the original result. *Therefore, electron-hole scattering is the dominant scattering and thermalization process.* Our results show that time-dependent, statically screened carrier-carrier Coulomb scattering provides an adequate description of carrier-carrier scattering processes in our case. The fit in Fig. 2(a) is not affected by a change in the hole density

from 1 to 4×10^{10} cm^{-2}, consistent with the conclusion above that hole-hole interactions do not play a major role. Changing the effective mass of the holes also has little effect on the cooling and thermalization; increasing the mass increases the scattering rate but decreases the scattering angle so that more collisions are elastic. Optical-phonon emission is found to be less important than electron-hole scattering for the energy-loss process for holes, except the transfer of the light holes to the heavy-hole band occurring in the first 100 fs after the pulse peak.[18]

The calculated picosecond time evolution also reproduces the experimentally observed decrease of the slope after 1 ps. However, EMC simulation calculates smaller luminescence intensity at long delays [Fig. 2(b)]. This result is most likely because the calculated carrier temperature at 7 ps is 40 K whereas that determined by fitting the spectrum is 30 K (Fig. 1). The calculated intensity at 7 ps would be given by the cross in Fig. 2(b), in good agreement with the experimental results, if the calculated carrier temperature were 30 K. The MC simulations show no change in the carrier temperature at 7 ps when we include dissipative acoustic-phonon scattering. Therefore, some other mechanism, possibly coupled plasmon-phonon scattering,[19] must be considered for more efficient cooling of the plasma in this range. Overall, the experimental and MC simulation results show a slow thermalization of heavy holes. The simulation shows that this suppression of cooling and, hence, the electron-hole scattering is due to the degeneracy of the electron system.

In conclusion, we have presented a study of initial hole relaxation in GaAs quantum wells. A judicious choice of sample and excitation parameters allows us to be in a regime where hole relaxation dominates the experimental results. Our results show that holes are *nonthermal* for nearly 800 fs after photoexcitation. This is in agreement with the results obtained by Monte Carlo simulations. Comparison with Monte Carlo simulations shows that hole-electron scattering is the dominant scattering mechanism, hole-hole scattering is not important, and that the holes remain nonthermal because electron degeneracy reduces this scattering rate. Statically screened Coulomb interaction provides an adequate description of hole-electron scattering in this case. Our results also show that spectral fit over a limited energy range should not be used to draw conclusions about the entire distributions.

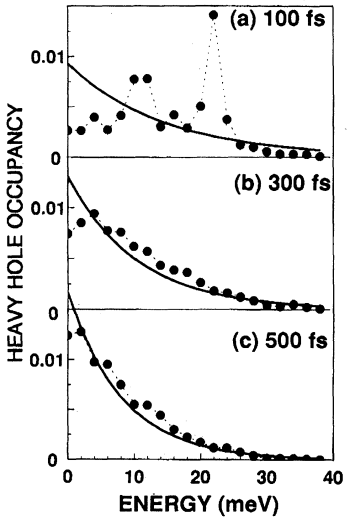

FIG. 3. Heavy-hole occupancy as a function of hole energy calculated from Monte Carlo simulation (circles). The occupancy is calculated for the time of (a) 100 fs, (b) 300 fs, (c) 500 fs. The curves show the thermalized (Maxwell-Boltzmann) distribution with the average energy calculated by EMC. The relative heights of the curves are correct, but the absolute value is arbitrary.

We thank T. C. Damen and DaiSik Kim for helpful discussions. S.L.C. would like to acknowledge the support of the ONR (Grant No. N00014-90-J-1821). S.M.G. would like to acknowledge the partial support of the Alexander von Humboldt Foundation for this work.

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