Time-Resolved Photoluminescence of Two-Dimensional Hot Carriers in GaAs-AlGaAs Heterostructures

J. F. Ryan, R. A. Taylor, A. J. Turberfield, and Angela Maciel
Clarendon Laboratory, Oxford University, Oxford, England

and

J. M. Worlock[(a)]
Bell Communications Research, Inc., Holmdel, New Jersey 07733

and

A. C. Gossard and W. Wiegmann
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
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We have studied the picosecond time dependence of luminescence from a two-dimensional electron system following absorption of an ultrashort light pulse. From our measurements we determine the temporal evolution of the carrier temperature, finding that the cooling of hot carriers is suppressed by a factor 60 below that predicted on a three-dimensional nondegenerate-electron model. Additionally, we determine the electron-hole radiative lifetime and invoke a hole trap to explain shortened luminscence lifetimes at low carrier densities.

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We report the first measurements of time-resolved photoluminescence from modulation-doped multiquantum well (MQW) semiconductor heterostructures. Our measurements, having a time resolution of $\simeq 20$ psec, allow us to observe (a) the hot-carrier cooling rates (electron-lattice interaction) and (b) the electron-hole recombination rates (radiative and nonradiative) as a function of both carrier temperature and initial carrier concentration. Hot-carrier behavior is extremely important in small, fast semiconductor devices and the radiative transitions are important in all photonic devices. Additionally, there is much interest attached to these fundamental interactions, especially in systems of reduced dimensionality.

The last few years have witnessed a strong increase in interest in two-dimensional (2D) carriers in semiconductors, confined either at single heterojunction interfaces or in quantum well structures.[1] In the GaAs-$Al_xGa_{1-x}As$ system, and especially in samples grown by molecular beam epitaxy (MBE), the smoothness and perfection of the interfaces[2] and the development of modulation doping[3] have led to electrons and holes with very high mobilities,[4,5] giving rise to extremely fast devices,[6,7] as well as to new phenomena, such as the fractional quantized Hall effect.[8]

Recent photoluminescence experiments on undoped MQW's have shown that exciton recombination is the dominant radiative process, and exciton lifetimes $\lesssim 1$ ns are measured.[9,10] In contrast, analyses of steady-state photoluminescence of modulation-doped GaAs-AlGaAs MQW[11] have demonstrated (1) that the dominant radiative processes are intrinsic band-to-band electron-hole recombination, (2) that the 2D electrons can be easily heated well above the lattice temperature either by electric currents or by excess photon energy, (3) that electron temperature (T_e) remains well defined even when T_e is on the order of 100 K above the lattice temperature (T_L), and (4) that the dominant process for energy loss by the entire electron system (for $T_e \gtrsim 50$ K) is LO-phonon emission. These we take as established facts, and our experiments are designed to measure directly the rates at which these processes take place.

Luminescence was excited using mode-locked pulses from a synchronously pumped dye laser. We used laser excitation at $h\nu = 2.03$ eV which is absorbed both in the GaAs wells and in the $Al_{0.23}Ga_{0.77}As$ layers (energy gap $\simeq 1.82$ eV). The luminescence was spectrally filtered and dispersed in a triple-grating spectrometer, and temporally dispersed by means of a Hadlands synchronously scanning streak camera locked to the 76-MHz laser pulse train. The best time resolution that can be obtained with the system is $\simeq 10$ ps, limited by the combined jitter of the laser and the driving electronics of the camera. In practice the resolution was $\simeq 20$ ps, determined by the width of the spectrometer slit. The spectral response of the photocathode is S20-like and the low-energy detection limit lies at ~ 1.5 eV.

The maximum laser power absorbed in the sample was $\simeq 50$ mW. The photoexcited carrier density was varied from 10^{15} cm^{-3} to 5×10^{18} cm^{-3} by use of different lenses and neutral-density filters. In each measurement the spot size on the sample was confirmed with calibrated pinholes. The carrier density quoted is the absorbed photon fluence divided by the penetration depth.

The steady-state photoluminescence from this sample at low temperatures shows a strong band peaking at 1.516 eV, which is near the band gap, E_g, of the GaAs layers. A shoulder toward high energy, at 1.528 eV, indicates the position of the Fermi level E_g+E_F. Luminescence in this range of energies stems from recombination of the degenerate electrons (Fermi-Dirac) with photoexcited holes (Maxwell-Boltzmann). The intensity in the tail of the band, $E > E_\text{F}+E_g$, arises from recombination of hot carriers, whose temperatures can be inferred from the exponential energy dependence of the intensity:

$$I(E) \propto \exp[-E/k_\text{B}T_e].$$

In the present experiments, we measure the temporal evolution of luminescence following pulsed excitation, to study both the cooling of the hot carriers and the recombination processes. Figure 1 shows time-resolved intensities at different energies, for the case of very strong pumping: $n=5\times10^{18}$ cm^{-3}. At $E=1.576$ eV, the measured rise and fall times are instrumental. At lower energies a rise time is resolved, but more dramatic is the large increase in lifetime. At $E=1.523$ eV (electrons from near bottom of conduction band) the measured lifetime is $\simeq 750$ ps, close to the lifetime in bulk GaAs ($\simeq 1$ ns). Here also the rise time is much longer, $\simeq 100$ ps.

Fitted lifetimes for energies on the entire luminescence band are given in Fig. 2. The very short lifetimes at high energies are evidence for rapid cooling of the 2D carriers. The rise times also give evidence of cooling, as the carriers populate the lower-energy states. In order to study the cooling process quantitatively, we measure $I(E)$ in the exponential tail at various delay times following excitation. At ~ 20-ps delay, T_e is well established, showing that electron-electron collisions are faster than LO-phonon emission. Figure 3 shows T_e versus time for the highest initial carrier density, $n=5\times10^{18}$ cm^{-3}. T_e (20 ps) is 105 K, although the initial T_e is certainly much higher, and substantial cooling has already occurred. After 150 ps, T_e has dropped to $\simeq 65$ K, and there is very little further cooling, out to several hundred picoseconds. For the lower excitation densities, the corresponding temperatures and cooling rates are lower.

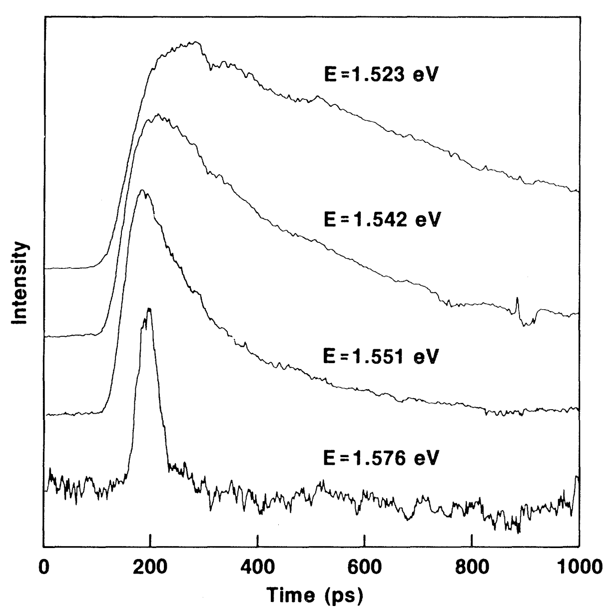

FIG. 1. Time-resolved hot luminescence from GaAs-AlGaAs MQW No. 8-4-81/2: $d_\text{superlattice}=774$ Å, $d_\text{GaAs}=258$ Å, $n_0=5\times10^{11}$ cm^{-2}. Laser energy $h\nu = 2.03$ eV. Photoexcited carrier density $=5\times10^{18}$ cm^{-3}. $T_L=4$ K.

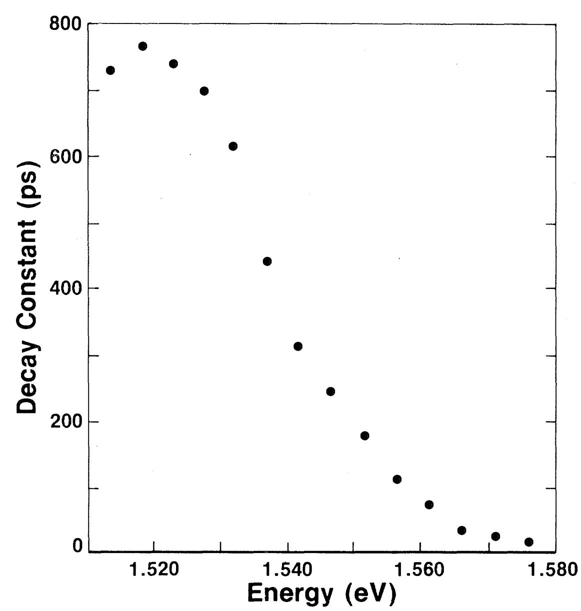

FIG. 2. Hot luminescence lifetimes vs energy for GaAs-AlGaAs MQW. Conditions as in Fig. 1.

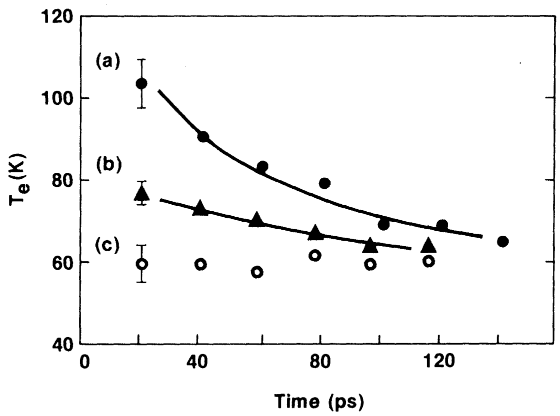

FIG. 3. Time depedence of carrier temperatures in GaAs-AlGaAs MQW for excited carrier densities (curve *a*) $n = 5\times 10^{18}$ cm^{-3}, (curve *b*) $n = 6\times 10^{16}$ cm^{-3}, (curve *c*) $n = 9\times 10^{14}$ cm^{-3}. The solid lines are obtained from Eq. (1) with $\tau_0 = 7$ ps, and specific heat $= nk_B$.

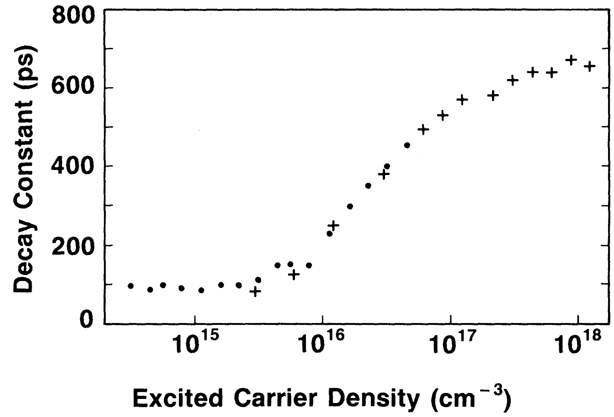

FIG. 4. Total luminescence (i.e., energy-integrated) lifetime vs excited carrier density for GaAs-AlGaAs MQW.

The solid lines in Fig. 3 show our calculated cooling curves, obtained with a time-honored formula for energy loss rate of an electron gas through LO-phonon emission,[12–14]

$$P(t) = (E_{LO}/\tau_0)\exp[-E_{LO}/k_B T_e(t)]. \qquad (1)$$

P is the mean power lost per carrier, and together with the specific heat determines the cooling curve. The LO phonon energy is $E_{LO} = 36.8$ meV. The exponential factor is the fractional number of carriers in a nondegenerate gas that can relax by LO-phonon emission, i.e., have energy $E > E_{LO}$. The expression (1), when fitted to experimental data, yields a value for τ_0 which is the time constant for the relaxation process. Our experiments require a value of 7 ps. The theoretical value of τ_0 is 0.11 ps in GaAs when the electron-phonon interaction is unscreened; however, a comparison of our result with other experimental determinations may be more meaningful. In bulk (3D) GaAs, with low-density excitation, von der Linde and Lambrich[14] found $\tau_0 = 0.12$ ps. At a somewhat higher carrier density, $n \sim 3\times 10^{17}$ cm^{-3}, Leheny *et al.*[15] give evidence for a scattering rate reduced by a factor of 5 below the low-density rate. Similarly, Shank *et al.*[16] found a reduced cooling rate in highly excited MQW structures, but the cooling rates were similar to those of the bulk. In all of these experiments temperatures and carrier densities were inferred from measurements of gain and absorption near the band edge. With $n \sim 2.5\times 10^{17}$ cm^{-3}, their measurements imply $\tau_0 \sim 0.7$ ps, while at lower densities the cooling is faster. The curves shown in Fig. 3 span the range from the low to high density, and yet we get a constant large value for τ_0, more than an order of magnitude larger than either the 3D or the 2D case.

There are several possible origins of this weakened electron-phonon interaction, which we shall discuss briefly. (1) The 2D plasma is efficient in screening the electron-phonon interaction. Indeed, because the relevant LO phonons have wavelengths, $2\pi/k$, shorter than the superlattice period, d, the 2D plasma frequency $[2n_s e^2 k/m^* e]^{1/2}$ is enhanced above the effective equivalent 3D plasma frequency $[4n_s e^2/m^* ed]^{1/2}$ by the factor $(kd/2)^{1/2}$. We estimate $kd/2 > 5$ for all important phonon processes in our experiments. Much more work is required here, including the effects of Landau damping and additional plasma dispersion. (2) Because of the reduced dimensionality the phonons that interact with the confined electrons may behave differently from those of the bulk.[17] The strength of the electron-phonon interaction may itself be reduced; alternatively the relaxation of the nonequilibrium phonons generated by the hot carriers may be substantially slower, so producing a bottleneck for the carrier–LO phonon scattering. (3) Electron degeneracy can be unexpectedly effective in denying access to lower-energy states to which electrons might be scattered.[18] (4) The apparent change of carrier temperature in time will depend on the wave vector dependence of the recombination processes. If low-energy carriers are preferentially depleted in either the radiative, or, more likely, nonradiative recombination, then the carrier temperature is increased in the process. In addition, carriers can be trapped into the wells from the barriers and their excess energy goes to raise the temperature. Further experimental and theoretical work is required in order to assess the importance of these various possibilities.

Let us return now to the luminescence emitted by already cooled electrons, near the steady-state peak. We saw in Fig. 2, for the highest initial carrier density, a luminescence lifetime of 750 ps. Somewhat surprisingly, we find that as we reduce the density, this lifetime decreases. The results are summarized in Fig. 4 where the mean lifetimes (obtained from energy-integrated spectra) are plotted against photoexcited carrier density. For $n \lesssim 5 \times 10^{15}$ cm^{-3} the lifetime is approximately constant at 100 ps. At higher densities the lifetime increases and it appears to saturate at about 700 ps for $n \gtrsim 10^{18}$ cm^{-3}. We find also that the radiative efficiency increases by a similar factor over the same density range. Furthermore, we observe at 1.503 eV a long-lived luminescence peak ($\tau \sim 1$ ns) which is resolved only at low excitation density and whose intensity saturates with increasing density. This set of observations provides clear evidence for efficient carrier trapping processes in MQW's. We suppose that there must be a shallow hole trap operating at low density so as to give a mean hole lifetime of $\simeq 100$ ps. At high densities these traps are saturated, and we see that the recombination time rises to a plateau at $\simeq 700$ ps, which we take to be the radiative lifetime. Carbon is known to be the predominant acceptor impurity in MBE-grown GaAs,[19] and so it may be the culprit. The fact that the binding energy is $\simeq 10$ meV indicates that the trap is located near the interface.[20]

These first experiments on time-resolved luminescence from 2D electron systems represent the most detailed investigations to date on hot-carrier cooling in semiconductors. They show that cooling is much slower than in the equivalent 3D case. It is suggested that future theoretical work should investigate the roles of screening, degeneracy, and reduced dimensionality in this reduction of the electron-phonon interaction. Further, these experiments have given us the intrinsic electron-hole radiative recombination lifetime, while suggesting the existence of a hole trap which shortens carrier lifetimes at low densities.

[a]Part of this work performed while at Bell Laboratories, Holmdel, N.J. 07733.

[1]See the Proceedings of the International Conferences on Electronic Properties of Two-Dimensional Systems, Surf. Sci. **98** (1980), and **113** (1983), and **141** (1984), etc.

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