Comparison of hot-carrier relaxation in quantum wells and bulk GaAs at high carrier densities

W. S. Pelouch, R. J. Ellingson, P. E. Powers, and C. L. Tang
Materials Science Center, Cornell University, Ithaca, New York 14853

D. M. Szmyd and A. J. Nozik
Solar Energy Research Institute, 1617 Cole Boulevard, Golden, Colorado 80401
(Received 4 September 1991)

An investigation of the hot-carrier relaxation in GaAs/(Al,Ga)As quantum wells and bulk GaAs in the high-carrier-density limit is presented. Using a time-resolved luminescence up-conversion technique with ≤ 80-fs temporal resolution, carrier temperatures are measured in the 100-fs-to-2-ns range. Our results show that the hot-carrier cooling rates in the quantum wells are significantly slower than in the bulk for carrier densities greater than 2×10^{18} cm^{-3}. A comparison is made with previous publications to resolve the confusion concerning the difference in cooling rates in quasi-two- and three-dimensional systems.

The investigation of hot-carrier relaxation in both quantum-well (QW) and bulk-semiconductor structures has been a topic of considerable interest.[1] It is well known that the hot-carrier cooling rate decreases with increasing carrier concentration in both QW and bulk structures. The first study comparing GaAs/(Al,Ga)As QW's and bulk GaAs reported similar cooling rates at a carrier density (n) of 2.5×10^{17} cm^{-3}.[2] Subsequent studies reported a much slower cooling rate in QW's than in bulk at higher carrier densities ($n > 10^{18}$ cm^{-3}).[3-6] Nevertheless, in a series of recent publications Leo *et al.*[7,8] have cited these previous results as being contradictory and concluded that the cooling rates of bulk GaAs and GaAs/(Al,Ga)As QW's are equivalent. These conclusions, however, were based on comparisons limited to the carrier density range of $10^{15} < n \leq 10^{18}$ cm^{-3}, generalizing that the independence of carrier cooling with dimensionality is also independent of carrier density. The relative hot-electron cooling rates for bulk and QW structures is an important, basic question that affects many applications of QW structures[9] and has inspired a large number of theoretical studies.[10] We have therefore reexamined this problem in the carrier density range of 2×10^{18} to 1.5×10^{19} cm^{-3} to resolve this apparent controversy.

The previous comparisons were made by means of nonlinear intensity correlation measurements (absorption, gain, luminescence) or time-resolved luminescence measured using a streak camera (with temporal resolution of $\sim$20 ps). Our present measurements are made using a time-resolved luminescence up-conversion technique[11-13] with a temporal resolution of $\sim$80 fs. Photoluminescence (PL) spectra were measured in the 100-fs-to-2-ns range from which effective electron temperatures and Fermi levels were deduced. We find a significantly slower hot-electron cooling rate for GaAs/(Al,Ga)As QW's than for bulk GaAs at carrier densities greater than 2×10^{18} cm^{-3}, the difference becoming more pronounced with increasing photogenerated carrier density. This difference could not have been observed in the carrier density range ($n < 10^{18}$ cm^{-3}) studied by Leo *et al.* or Shank *et al.*[2]

The samples were grown by metalorganic chemical-vapor deposition on $\langle 100 \rangle$ GaAs substrates. The bulk samples consist of (i) a 2000-Å and (ii) a 4000-Å nominally undoped GaAs layer surrounded by thin (<0.2 μm) (Al,Ga)As layers which are transparent to the 1.97-eV pump. The multiple-quantum-well (MQW) structures consist of fourteen periods of 135-Å thick GaAs and 400-Å-thick $Al_{0.48}Ga_{0.52}As$ grown on the GaAs substrate. The GaAs layers in the MQW contain a 3.5% Al concentration which raises the band gap by $\sim$50 meV and ensures that the luminescence signal of the high-energy tail of the MQW is not confused with that of the GaAs substrate.

Time-resolved luminescence spectra were obtained by the technique of sum frequency generation,[11,12] using an arrangement nearly identical to that of Ref. 13 with the following modifications: the luminescence was collected in reflection; a 1.35-mm barium metaborate (BBO) crystal was used for up conversion; and a cooled, low-noise (0.5-Hz dark count) Bi photomultiplier tube was used for detection, which improves the dynamic range. The group-velocity mismatch between the pump and the luminescence wavelengths is 50 fs/mm in BBO and the pump pulse duration is ≤ 50 fs, yielding an overall temporal resolution of ≤ 80 fs. We calculate[14] the spectral resolution to be 10 meV.

Time-resolved luminescence spectra were recorded at room temperature at the following incident pump powers: 25, 12.5, and 5 mW. The PL spectra of the 2000-Å bulk GaAs sample contain a contribution from the substrate at the earliest times (≤ 5 ps) until the substrate carriers have diffused sufficiently. Thus, results from this sample are discussed only for delays ≥ 5 ps. The bulk GaAs PL spectra presented below correspond to the 4000-Å sample since the substrate absorption is minimal. Representative spectra and carrier temperature fits for the 4000-Å bulk GaAs sample at each excitation power are shown in Fig. 1 for several time delays. All spectra were corrected for the spectral response of the

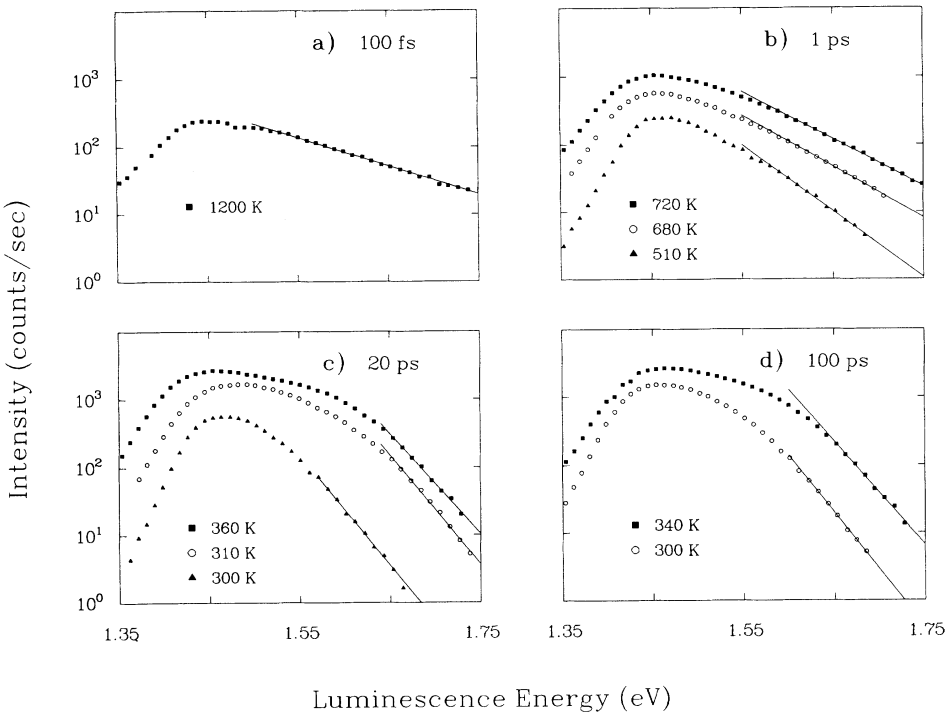

FIG. 1. Representative time-resolved luminescence spectra for the 4000-Å bulk GaAs sample at room temperature at each excitation power (squares, 1×10^{19} cm^{-3}; circles, 5×10^{18} cm^{-3}; triangles, 2×10^{18} cm^{-3}). The carrier temperature fits (straight lines) and values are shown for each carrier density at delay times of (a) 100 fs, (b) 1 ps, (c) 20 ps, and (d) 100 ps.

up-conversion system. The electron temperatures were determined by fitting the high-energy tails of the spectra; only the region which is linear on a semilogarithmic plot was chosen for the fit. Figure 1(a) displays a plot of the PL spectrum at 100 fs for the highest carrier density that shows that the initial carrier temperature is 1200 K. Figures 1(b)–1(d) show the evolution of the carrier distribution at time delays of 1, 20, and 100 ps for each excitation power. The quasi-Fermi energy of the electron distribution was deduced by fitting the PL spectra and is located approximately at the position of the kink in the semilogarithmic plot of the spectra [see Figs. 1(c) and 1(d)]. The carrier density (n) was calculated from the values of the quasi-Fermi levels at the highest excitation power and then scaled according to excitation power. The value of the carrier density agrees well with our calculation using beamwaist, reflection, and absorption parameters; we estimate this value to be accurate to within 15%. The carrier densities for the 4000-Å GaAs sample are 1×10^{19}, 5×10^{18}, and 2×10^{18} cm^{-3}, corresponding to the incident excitation powers of 25, 12.5, and 5 mW, respectively. The carrier densities are 45% greater for the 2000-Å GaAs sample (PL spectra not shown).

Similarly, spectra for the MQW sample were recorded at the same pump powers as the bulk. Representative spectra and carrier temperature fits are presented in Fig. 2 for each excitation power. The peak at 1.43 eV corresponds to the substrate luminescence and the following peaks are attributed to the QW subbands. A few percent of the electrons in the MQW have energies that exceed the height of the barrier potential which can be observed as a sudden drop in the PL intensity at energies >1.76 eV. The fraction of energy beyond 1.76 eV can be calculated by extrapolating the high-energy PL tail from the carrier temperature fit and integrating the MQW PL spectra (after subtracting the bulk spectra). The average energy of these unbound carriers is about twice as great as those in the QW region. At 1 ps this fraction is calculated to be $<4\%$ of the energy ($<2\%$ of the carriers) at the highest carrier density, $<2.5\%$ of the energy ($<1.2\%$ of the carriers) at the medium carrier density, and negligible at the lowest carrier density. The possible effects of this on the hot-carrier cooling is discussed in more detail below.

The absorption coefficient at the pump wavelength in the MQW sample was measured on an etched sample and is slightly less than that of GaAs due to the 3.5 at. % Al doping. The MQW carrier densities are calculated to be 35% *greater* than the 4000-Å bulk GaAs sample and 6% *less* than the 2000-Å GaAs sample for equal pump powers. Line-shape analysis of the MQW spectra is somewhat more difficult due to the presence of several subbands, but the measured quasi-Fermi levels yield carrier densities that agree well with the above calculations.

In Fig. 3 we compare the carrier temperatures of each sample versus the initial carrier density for several time delays. At 1 ps [Fig. 3(a)] the bulk and MQW samples have similar carrier temperatures and an identical dependence on carrier density. In the 5–100-ps range [Figs. 3(b)–3(d)] the carrier temperatures of the MQW sample remain much hotter than the bulk samples and show a substantially different dependence on initial carrier density, as seen by the different slopes of the lines. In this temporal regime, the reduction in hot-carrier cooling is greatly enhanced in the MQW structure in comparison to the bulk as the carrier density increases. The carrier temperature of the 4000-Å bulk GaAs sample at the greatest carrier density has cooled to 310 K by 2000 ps (not shown), which suggests that lattice heating is not significant. Since both the bulk GaAs and the MQW samples have approximately the same capping layer and substrate material, any lattice heating effects are nearly

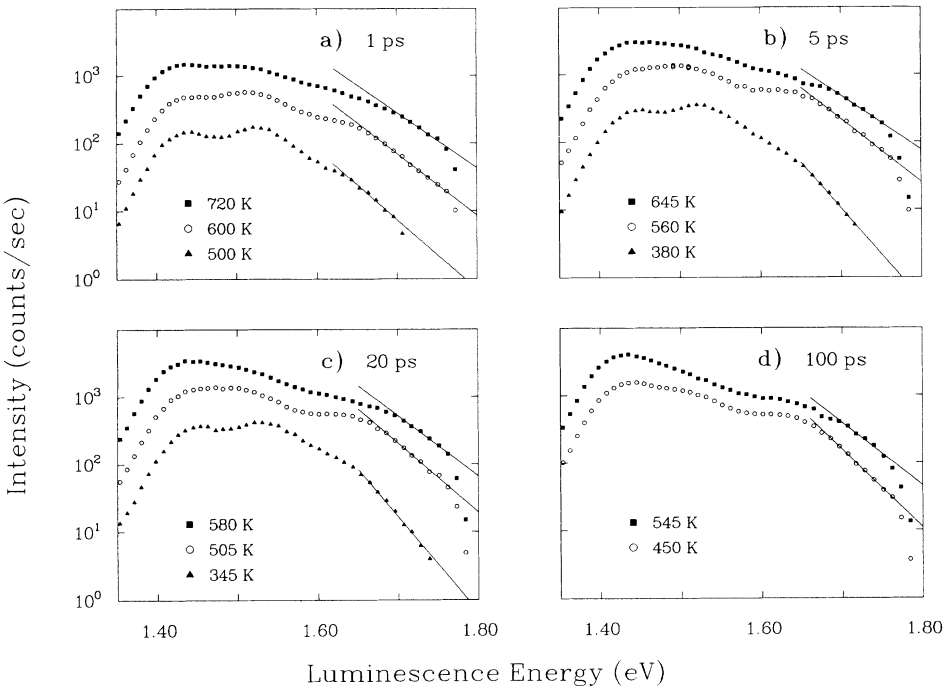

FIG. 2. Representative time-resolved luminescence spectra for the MQW sample at room temperature at the same excitation power as the bulk (squares, 1.4×10^{19} cm^{-3}; circles, 6.8×10^{18} cm^{-3}; triangles, 2.7×10^{18} cm^{-3}). The carrier temperature fits (straight lines) and values are shown for each carrier density at delay times of (a) 1 ps, (b) 5 ps, (c) 20 ps, and (d) 100 ps.

identical.

It must be noted that the carriers in the MQW sample are excited only within the well region, but at energies above the conduction band ($\sim$0.5 eV) that exceed the height of the barrier potential ($\sim$0.3 eV). Since the holes are bound within the quantum well at all times, the scattering rate or diffusion of electrons to the barrier region is reduced due to Coulomb attraction.[15] The electrons are bound in the QW extremely rapidly due to phonon emission and form a hot Maxwellian distribution quicker than can be resolved by experiment (<100 fs). Any carriers that reach the barrier region would return to the QW within a few picoseconds[15,16] and would not affect the PL spectra for times ≥ 20 ps. During the hot-carrier relaxation a few percent of the electrons have energies that exceed the height of the barrier potential (see Fig. 2 and above discussion), which also may affect the cooling rates in the MQW sample. If all these carriers were suddenly "dumped" in the QW region we calculate a slight rise in the Fermi level and an increase in carrier temperature of about 25 K (15 K) at the highest (medium) carrier density. Therefore it is unlikely to expect that the effects discussed above could account for the tremendous increase in the QW hot-carrier relaxation time which occurs on a time scale of hundreds of picoseconds with carrier temperatures of up to 200 K above the bulk.

The mechanism that is generally considered as being

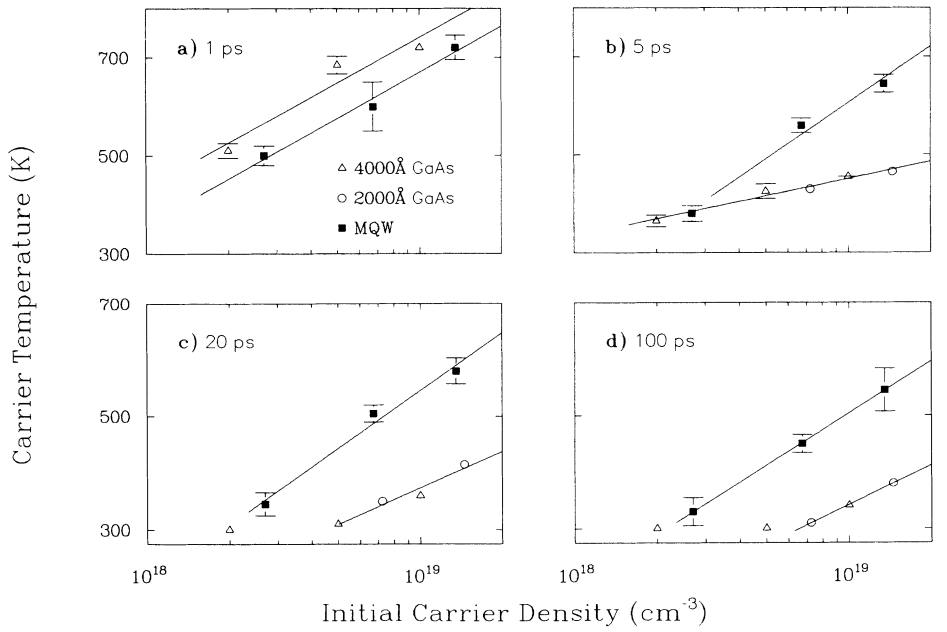

FIG. 3. A comparison of the carrier temperatures vs initial carrier density for the 4000-Å bulk GaAs (triangles), the 2000-Å bulk GaAs (circles), and the MQW (squares) samples at time delays of (a) 1 ps, (b) 5 ps, (c) 20 ps, and (d) 100 ps. The straight lines are guides to the eye. The different dependence in carrier cooling with respect to initial carrier density between the MQW and bulk samples is most evident at intermediate times (5–20 ps) and high carrier densities ($n>2\times10^{18}$ cm^{-3}).

responsible for the slowed electron cooling in semiconductors is the generation of a nonequilibrium phonon distribution (hot phonons) and subsequent reabsorption of the phonons by the carriers which prolong the hot-carrier relaxation time. Recent analyses by Ge *et al.*[17] and Das Sarma[18] have shown that the relaxation time is mostly affected by the hot-phonon lifetime and the carrier density rather than just the electron-phonon interaction. In both these treatments it is reported that the hot-phonon effect is enhanced in quantized structures due to two-dimensional (2D) confinement and becomes more pronounced with increasing carrier density. Furthermore, the hot-carrier relaxation time is calculated to be faster in structures with less confinement, i.e., thicker QW and bulk structures.

These data show that the hot-carrier cooling is different in QW and bulk GaAs at higher carrier densities ($n > 10^{18}$ cm^{-3}). From an experimental point of view, there is little or no contradictory evidence in the comparison of quasi-2D and -3D hot-carrier cooling rates. The confusion arises in the interpretation of experiments that were carried out in the low- to mid-carrier density range ($n < 10^{18}$ cm^{-3}) where the cooling rates appear to be the same in both bulk and MQW structures. The experiment described in this Brief Report represents a significant improvement over the previous methods in the ability to characterize the evolution of the hot-carrier distribution. Unlike nonlinear intensity correlation experiments,[2,3] the state of the system can evolve without perturbation after the pump (i.e., there is no probe). The method of luminescence detection using a streak camera[7,8] lacks the ability to detect hot luminescence for times less than or on the order of the phonon lifetime due to the limited temporal resolution which requires the experiment to be carried out at low temperatures in order to observe any effects of the hot-carrier relaxation. The results of these previous studies agree with our observations *in their respective carrier-density ranges.* The similarity in the cooling rates of MQW and bulk GaAs at $n = 2\times10^{18}$ cm^{-3}, as seen in Fig. 3, suggests that these cooling rates would be nearly identical at lower carrier concentrations in agreement with the observations of Leo *et al.* It should also be noted that we have observed that the precise values of the carrier temperatures of a variety of MQW samples depend on both sample quality and age [the latter presumably due to degradation of the samples with oxidation of the (Al,Ga)As surface], but the qualitative results of this study are not affected.

In conclusion, the hot-carrier cooling rates in GaAs/(Al,Ga)As quantum wells and bulk GaAs have been investigated in the high-carrier-density limit ($n \geq 2\times10^{18}$ cm^{-3}). The cooling rates of the quantum-well structure are significantly slower than that of the bulk for $n \geq 5\times10^{18}$ cm^{-3} and similar at $n = 2\times10^{18}$ cm^{-3}, in agreement with the results of previous publications. The investigation of hot-carrier relaxation in the high-carrier-density limit is important in order to understand the physics governing quasi-2D and -3D hot-carrier relaxation over the widest range of conditions and may have important consequences in the development of semiconductor devices.

The research at Cornell was supported by the Joint Services Electronics Program and the National Science Foundation. A.J.N. and D.M.S. were supported by the U.S. Department of Energy, Divison of Basic Energy Sciences/Chemical Sciences.

[1]J. Shah, Solid State Electron. **32**, 1051 (1989), and references therein.
[2]C. V. Shank, R. L. Fork, R. Yen, J. Shah, B. I. Greene, A. C. Gossard, and C. Weisbuch, Solid State Commun. **47**, 981 (1983).
[3]Z. Y. Xu and C. L. Tang, Appl. Phys. Lett. **44**, 692 (1984).
[4]H. Ukichi, Y. Arakawa, H. Sakaki, and T. Kobayashi, Solid State Commun. **55**, 311 (1985).
[5]S. A. Lyon, J. Lumin. **35**, 121 (1986).
[6]A. J. Nozik, C. A. Parsons, D. J. Dunlavy, B. M. Keyes, and R. K. Ahrenkiel, Solid State Commun. **75**, 297 (1990).
[7]K. Leo, W. W. Rühle, and K. Ploog, Phys. Rev. B **38**, 1947 (1988); Solid State Electron. **32**, 1863 (1989).
[8]K. Leo, W. W. Rühle, H. J. Queisser, and K. Ploog, Appl. Phys. A **45**, 35 (1988); Phys. Rev. B **37**, 7121 (1988).
[9]See, for example, *High Speed Semiconductor Devices,* edited by S. M. Sze (Wiley, New York, 1990).
[10]See, for example, P. Lugli, P. Bordone, S. Gualdi, P. Poli, and S. M. Goodnick, Solid State Electron. **32**, 1881 (1989).
[11]H. Mahr and M. D. Hirsch, Opt. Commun. **13**, 96 (1975).
[12]T. C. Damen and J. Shah, Appl. Phys. Lett. **52**, 1291 (1988).
[13]F. W. Wise and C. L. Tang, Solid State Commun. **69**, 821 (1989).
[14]J. Shah, IEEE J. Quantum Electron. **QE-24**, 276 (1988).
[15]B. Deveaud, J. Shah, T. C. Damen, and W. T. Tsang, Appl. Phys. Lett. **52**, 1886 (1988).
[16]D. Bimberg, J. Christen, A. Stechenborn, G. Weimann, and W. Schlapp, J. Lumin. **30**, 562 (1985); D. J. Westland, D. Mihailovic, J. F. Ryan, and M. D. Scott, Appl. Phys. Lett. **51**, 590 (1987).
[17]W. Ge, Z. Xu, Y. Li, Z. Xu, J. Xu, B. Zheng, and W. Zhuang, J. Lumin. **46**, 137 (1990).
[18]S. Das Sarma, in Proceedings of the 7th International Conference on Hot Carriers in Semiconductors, Nara, Japan, 1991 [Semicond. Sci. Technol. (to be published)].