Hot carrier dynamics in a (GaInSb/InAs)/GaInAlAsSb superlattice multiple quantum well measured with mid-wave infrared, subpicosecond photoluminescence upconversion

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We have extended the technique of subpicosecond photoluminescence upconversion to the mid-wave infrared spectral region and have used this system to investigate the energy relaxation of hot, optically injected electron-hole pairs in a narrow-band-gap (2.32 μm) (GaInSb/InAs)/GaInAlAsSb superlattice multiple quantum well. These and similar structures are currently of interest as the active region for mid-wave infrared diode lasers. The measurements demonstrate that carriers, which are injected with nearly 1 eV of excess energy, are well described by a hot, thermalized distribution in the wells within 2 ps after excitation. For a carrier density of 10^{17} cm^{-3}, cooling by optical phonon emission is essentially complete 15 ps after injection. By fitting the time dependence of the carrier temperature, we estimate an effective carrier-optical-phonon scattering time of 1.2 ps. © *1997 American Institute of Physics.*
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The broad range of potential applications for compact and efficient sources of intense, mid-wave infrared (MWIR, 2–5 μm) coherent radiation has generated significant interest in the development of MWIR semiconductor laser diodes. Although there are several other candidate structures for such sources,[1] one promising system is based on strained superlattice multiple quantum wells composed of GaInSb/InAs type II, broken-gap superlattice segments with barrier layers composed of AlGaSb, GaInAsSb, or GaInAlAsSb alloys. Such structures, which have been implemented successfully in 3.3–3.9 μm lasers,[2,3] offer the potential for continuous-wave operation at noncryogenic temperatures and at wavelengths throughout the MWIR. Moreover, these materials offer great flexibility in design, thereby providing an opportunity for band structure engineering[4–7] to optimize device performance.

Although broken-gap superlattice multiple quantum wells are promising for MWIR laser diodes, current experimental information regarding the electronic properties of these structures is extremely limited. We have recently reported the application of subpicosecond MWIR pulses from an optical parametric oscillator[8] to measurements of Shockley-Read–Hall and Auger recombination in one such structure.[9] These nonradiative recombination processes play a critical role in the performance of diode lasers comprised of such structures. Another process that is closely linked to diode laser performance is the manner in which hot carriers injected into the active region relax to the band edge states. It is well known that hot-carrier effects can adversely impact diode laser performance[10] through reduced occupation of the band edge states, decreased carrier confinement and, perhaps most importantly for long wavelength lasers, enhanced Auger recombination. Furthermore, Auger recombination itself tends to heat the carrier distribution and can lead to prolonged energy relaxation.[11] Since the cooling of a hot carrier distribution is intimately related to carrier-optical-phonon scattering, the refinement of our understanding of this process in narrow band gap semiconductors may ultimately lead to improved MWIR diode lasers.

Here, we apply subpicosecond time-resolved photoluminescence upconversion[12] to the first study of energy relaxation of hot carriers through carrier-phonon scattering in the antimony-based broken-gap superlattice system. To our knowledge, this is also the first demonstration of subpicosecond photoluminescence upconversion in the MWIR. We extract the time-dependent carrier temperature from analysis of the hot luminescence and subsequently determine an average carrier-optical-photon scattering rate.

The structure studied is composed of 10 quantum wells, each comprised of a 6.5-period segment of a $Ga_{0.75}In_{0.25}Sb$/InAs (28.4 Å/6.7 Å) broken-gap superlattice, bounded by 335 Å $In_{0.18}Ga_{0.53}Al_{0.29}As_{0.17}Sb_{0.83}$ barriers. The outermost barrier layers are each 1000 Å thick. The structure was grown by molecular beam epitaxy on an undoped GaSb substrate. The 8 K band gap energy of the superlattice quantum well is 0.532 eV, and the quintarnary barrier band gap is estimated to be 1.13 eV. The 8 K time-integrated photoluminescence is shown in Fig. 1; the arrows indicate the energies at which subsequent time-resolved measurements were performed. Shown in the inset is the conduction/heavy-hole band alignment for the structure, along with the lower-lying confined conduction ($C1$–$C3$) and heavy-hole valence ($HH1$-$HH3$) band quantum well energy levels obtained from a superlattice $\mathbf{K}\cdot\mathbf{p}$ model.[13] The photoluminescence peak is associated primarily with $C1$-$HH1$ (0.532 eV) transitions, although we clearly also observe hot luminescence from $C2$-$HH2$ (0.577 eV) and $C3$-$HH3$ (0.643 eV) transitions in the time-resolved measurements.

The upconversion measurements were designed to pro-

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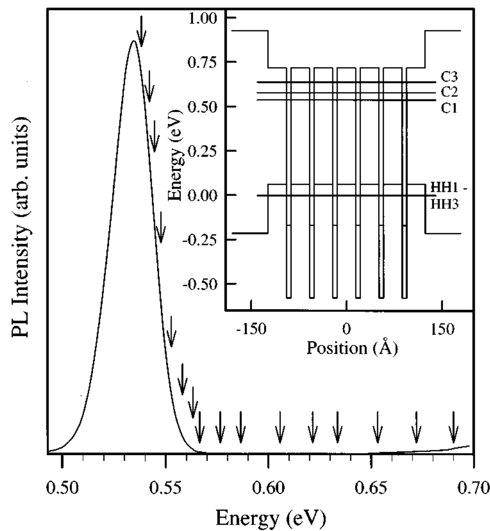

FIG. 1. Time-integrated photoluminescence at 8 K. The arrows indicate energies at which the time-resolved photoluminescence was measured. The inset illustrates the broken-gap superlattice quantum well conduction/heavy-hole band alignment and the three lowest confined electron ($C1$-$C3$) and heavy-hole ($HH1$-$HH3$) energy levels. Note that the latter are nearly degenerate and not resolved on the scale of this figure.

vide subpicosecond temporal resolution in the MWIR. We illuminated the sample with 140 fs pulses at 840 nm from a modelocked Ti:sapphire laser. For the data reported here, 10 mW of average power at 76 MHz was focused to a 50 μm spot on the sample surface, producing a carrier density of approximately 10^{17} cm^{-3} in the wells. The density was determined by fitting measured time-resolved photoluminescence spectra to calculated spectra. The density evaluated in this manner is in good agreement with an estimate based on the optical excitation density. The photoluminescence was collected and focused into a 1-mm-thick, 37°-cut, potassium titanyl arsenate (KTA) crystal, which is designed for noncollinear, type II, sum-frequency generation using 840 nm and MWIR radiation. A second gating pulse from the Ti:sapphire laser was focused onto the same spot on the KTA crystal at an angle of 16° with respect to the focused photoluminescence. The upconverted radiation was detected through a 1/3 m monochromator using photon counting instrumentation and a thermoelectrically cooled photomultiplier tube with a GaAs photocathode. The 200 fs temporal response of the system was determined by a cross correlation between the gating pulse and excitation light scattered from the sample surface, a measurement that also provides absolute timing between the gating and excitation pulses.

Time-resolved photoluminescence data are shown in Fig. 2 for five of the emission energies indicated in Fig. 1 (for clarity, not all the data are shown). For energies within the wells but far above the band edge, we observe a rapid transient emission as is evident in Fig. 2(a). For energies deeper in the well, the emission becomes more intense, the peak shifts to longer times, and the rise time increases. While these data indicate that most of the relaxation to the band edge states occurs within 15 ps of excitation, it is clear that carriers continue to accumulate in these states for more than 50 ps (see 0.539 eV data). The data are suggestive of an initial rapid cooling by optical phonon emission. As the carrier distribution temperature continues to decrease, the relaxation process slows significantly. This is expected since, for carrier temperatures well below the characteristic optical phonon temperature, cooling will occur only through optical phonon emission by carriers in the high energy tail of the distribution and through low energy acoustic phonon emission.[14–16] As illustrated by Fig. 2(b), the decay rate of the luminescence is even more strongly energy dependent. The decay is determined by optical phonon emission at the highest energies and by carrier recombination near the band edge. These data indicate a carrier recombination time of approximately 2.5 ns.

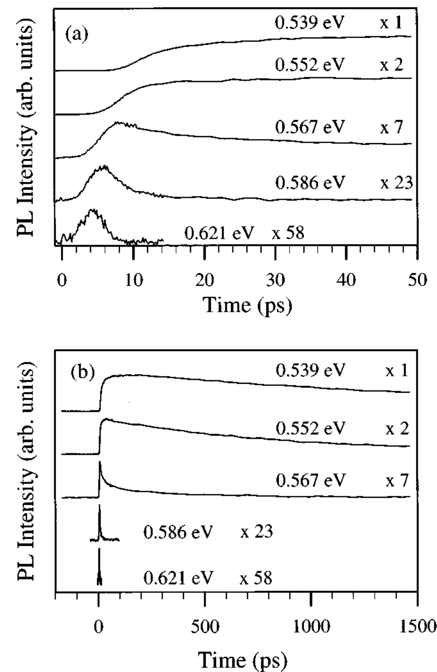

FIG. 2. Time-resolved photoluminescence at various energies emphasizing (a) the rise time and (b) the decay time of the emission. Note the differing intensities as given by the scaling factors.

To quantify the hot-carrier relaxation, we converted the data in Fig. 2 to time-resolved spectra, as shown in Fig. 3

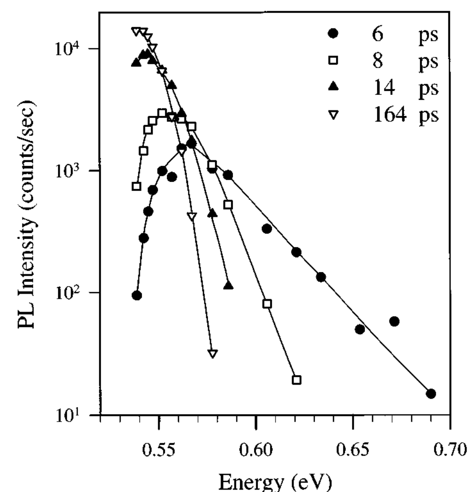

FIG. 3. Time-resolved photoluminescence spectra at various times after excitation. The solid curves are visual guides.

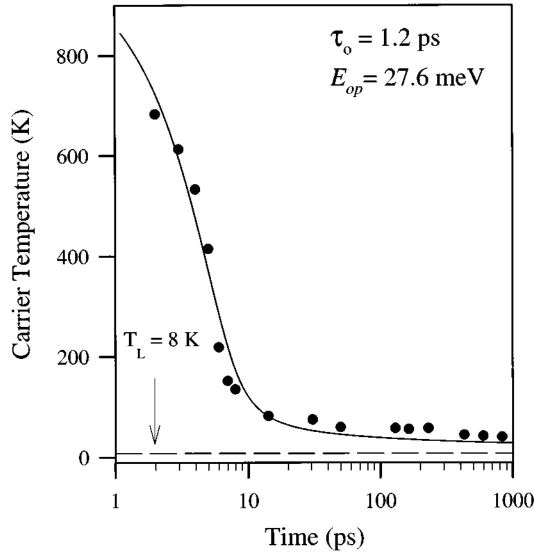

FIG. 4. Temporal dependence of the carrier distribution temperature. The dashed horizontal line indicates the lattice temperature and the solid curve is a theoretical fit based on Eq. (2) in the text. For the fit, an average optical phonon energy of 27.6 meV and an effective electron-optical-phonon scattering time of 1.2 ps were used.

(again, not all data are shown). We observe that, for time delays of 2 ps and beyond, the spectra exhibit exponential high energy tails, indicating that on such time scales the carrier distribution is thermalized. To extract the carrier temperature T_c, the high energy portion of these spectra were fit to[14]

$$I(E) \propto \alpha(E) E^2 \ \exp(-E/k_b T_c). \quad (1)$$

Here, $I(E)$ is the upconverted luminescence intensity at energy E, $\alpha(E)$ is the absorption coefficient computed from the superlattice $\mathbf{K}\cdot\mathbf{p}$ model, and k_B is Boltzmann's constant. As shown in Fig. 4, we find that the carrier temperature rapidly decreases from approximately 700 K at 2 ps after excitation to approximately 80 K at 15 ps after excitation. The cooling process then slows significantly, in agreement with our earlier qualitative discussion of the data in Fig. 2.

We use the time-dependent carrier temperature to estimate an effective carrier-optical-phonon scattering rate. With an optical phonon energy of $E_{\mathrm{op}}=27.6$ meV based on a weighted average of values[17] for the binary constituents of the wells, we fit the data in Fig. 4 with[18]

$$P(t) = -\frac{E_{\mathrm{op}}}{\tau_0 \sqrt{\pi/2}} \frac{e^{-x_c} - e^{-x_L}}{1 - e^{-x_L}} \left[\sqrt{x_c/2} e^{x_c/2} K_0(x_c/2) \right]. \quad (2)$$

Here, $P(t)$ is the mean power lost per carrier, τ_0 is an effective carrier-optical-phonon scattering time, K_0 is the modified Bessel Function of order zero, and $x_{c(L)} = E_{\mathrm{op}}/k_B T_{c(L)}$, with $T_{c(L)}$ the carrier (lattice) temperature. The fit to the data of Fig. 4, which is shown by the solid curve, yields $\tau_0 = 1.2$ ps. This scattering time is representative of all electron *and* hole energy relaxation processes. A theoretical estimate of the *electron-polar-optical-phonon* scattering time, $\tau_{\text{e-pop}}$, can be obtained from a simplified model that neglects hot-phonon effects and assumes a three-dimensional Frölich interaction with a nondegenerate carrier distribution.[18] We note that extension of this model to a two-dimensional system does not strongly influence the scattering time.[18] Using weighted averages for the static and high-frequency dielectric constants appropriate to the binary constituents of the superlattice[17] and using the electron effective mass of 0.0346 m_0 from the superlattice $\mathbf{K}\cdot\mathbf{p}$ band structure, we obtain $\tau_{\text{e-pop}}=0.48$ ps. This scattering time does not include contributions from hole-phonon scattering, and therefore $\tau_{\text{e-pop}}$ should provide only a *reasonable upper limit* for τ_0. Since it is well known that hot phonons increase the effective carrier-phonon scattering time,[14–16,18] our measured $\tau_0 > \tau_{\text{e-pop}}$ may indicate that hot phonons are influencing our results. We emphasize, however, that no definitive statement can be made in this regard without a detailed investigation of the density dependence of τ_0. Such studies are currently under way.

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