AIP | Applied Physics Letters

Suppression of intervalley scattering in Ga(As)Sb quantum wells

K. C. Hall, S. W. Leonard, H. M. van Driel, A. R. Kost, and E. Selvig

Citation: Applied Physics Letters **77**, 2882 (2000); doi: 10.1063/1.1322368
View online: http://dx.doi.org/10.1063/1.1322368
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/77/18?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS VOLUME 77, NUMBER 18 30 OCTOBER 2000

Suppression of intervalley scattering in Ga(As)Sb quantum wells

K. C. Hall,[a] S. W. Leonard, and H. M. van Driel
Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario M5S 1A7, Canada

A. R. Kost[b] and E. Selvig[c]
HRL Laboratories LLC, 3011 Malibu Canyon Road, Malibu, California 90265

(Received 16 March 2000; accepted for publication 6 September 2000)

Femtosecond time-resolved reflectivity was measured near the 1.55 μm absorption edge of several $GaAs_xSb_{1-x}/AlSb$ quantum well samples. On the basis of differences in the reflectivity recovery kinetics and plateau values, we deduce that $\Gamma-L$ intervalley scattering can be effectively suppressed for $x \geqslant 0.19$. This is consistent with calculations incorporating confinement and strain effects which give $L-\Gamma$ energy separations of 29 $(x=0)$ and 109 meV $(x=0.19)$. Suppression of intervalley scattering can lead to increased internal quantum efficiency and higher carrier mobility in 1.55 μm based devices. © *2000 American Institute of Physics.* [S0003-6951(00)04244-3]

Heterostructures based on GaSb and its alloys have become increasingly important due to potential applications in 1.55 μm communication devices.[1–3] Because of the small energy separation between Γ and L conduction band edges, however, phonon-assisted intervalley scattering plays a crucial role in these materials. Carrier occupancy of the L valleys is often undesirable since the higher L valley effective mass reduces carrier mobility and provides a higher density of states from which electrons can recombine with holes. L valley recombination reduces radiative quantum efficiency, impeding laser applications. The prevalence of intervalley scattering in GaSb and GaSb/AlSb multiple quantum wells was experimentally verified using femtosecond time-resolved reflectivity measurements by Smith *et al.*[4]

In this letter, we report the use of similar techniques to probe carrier dynamics in $GaAs_xSb_{1-x}/AlSb$ multiple quantum wells. The incorporation of As into GaSb increases the energy of L valley states relative to the Γ valley due to the larger $L-\Gamma$ separation in GaAs relative to its band gap. (The $L-\Gamma$ energy separation is 300 meV in GaAs, which is 21% of the band gap, compared to 61 meV in GaSb, corresponding to only 8% of the band gap energy.)[5] Elevation of L states relative to Γ substantially reduces their influence on band edge carrier dynamics and optical properties, thereby improving material characteristics for applications. From our measured band edge reflectivity kinetics, we deduce that for an As fraction (x) of 0.19 (and higher values) intervalley (IV) scattering can be made negligible for electrons with energy $\leqslant 3$ optical phonons ($\hbar\Omega_{IV}=26$ meV in GaSb). This conclusion is consistent with calculations of the $L-\Gamma$ energy separation in $GaAs_xSb_{1-x}/AlSb$ quantum wells as a function of As fraction.

The $GaAs_xSb_{1-x}/AlSb$ multiple quantum wells with As fractions between $x=0$ and 0.3 were grown by molecular beam epitaxy and designed to have a band gap energy of 0.8 eV (1.55 μm) at 295 K. The band gap energy as well as the energy separation between the L and Γ minima of the $n_z=1$ conduction subband were calculated taking into account As fraction, quantum confinement, and strain.[6] The calculated value of E_L-E_Γ is shown as a function of As fraction in Fig. 1. The $L-\Gamma$ valley separation exhibits a monotonic increase with As fraction. The model predicts a separation in the valley edges of 29 meV in GaSb/AlSb wells, indicating that nearly all carriers in the Γ valley will have access to L valley states through phonon absorption and emission processes.[7] However, a value for E_L-E_Γ of 100 meV occurs for the relatively small As fraction of 0.17. This separation, corresponding to $\sim 4\hbar\Omega_{IV}$, will induce a significant change in the band edge properties since intervalley scattering will be energetically prohibited for electrons within approximately 75 meV of the band edge.

We concentrate here on samples grown with $x=0$ and 0.19, for which the predicted $L-\Gamma$ energy separations are 29 and 109 meV, respectively, and the quantum well widths are 8 and 5.1 nm. Due to the presence of bowing in the valence bands of the ternary system Ga(As)Sb,[8] and because the quantum well layers experience tensile strain, confinement tuning of the band gap to 1.55 μm requires that the quantum well layer thickness be lowered with increasing As fraction

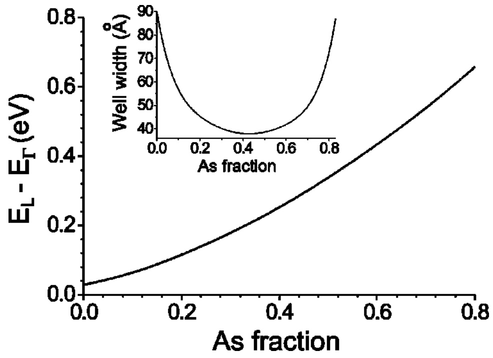

FIG. 1. Calculated energy separation between the Γ and L minima of the $n_z=1$ conduction subband in $GaAs_xSb_{1-x}/AlSb$ quantum wells. Inset: Quantum well width necessary to provide a band gap transition wavelength of 1.55 μm plotted vs As fraction, taking into account strain-induced subband edge shifting.

---

[a] Electronic mail: kchall@physics.utoronto.ca

[b] Present address: Optical Sciences Center, University of Arizona, 1630 East University Boulevard, Tucson, Arizona 85721.

[c] On leave from the Department of Physical Electronics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway.

for concentrations below 40%, as indicated in the inset of Fig. 1. The AlSb barrier layers are 8 nm thick for both samples. From photoluminescence experiments at 295 K, the band gap wavelengths were measured to be 1.55 μm ($x=0$) and 1.52 μm ($x=0.19$). The samples were fabricated as multiple quantum wells, containing 6 periods ($x=0$) or 60 periods ($x=0.19$), and grown on a Bragg mirror with a peak reflectivity of 80% at 1.55 μm. The mirror consists of five periods of $Ga_{0.68}Al_{0.32}Sb/AlSb$ $\lambda/4$ layers on a GaSb substrate.

Pump-probe reflectivity experiments were carried out at 295 K in a standard noncollinear geometry, employing pump and probe pulses with perpendicular linear polarizations. The apparatus used is similar to that of Ref. 1. The Bragg mirrors permitted the use of a reflection geometry in detecting pump-induced modulation of absorption. In this geometry, changes in quantum well absorption are linearly proportional to changes in probe reflectivity. The optical source is a 250 kHz repetition rate optical parametric amplifier, providing tunable 100 nJ, 115 fs pulses. For each sample, the pulse center wavelength (1.53 μm; $x=0$, 1.49 μm; $x=0.19$) was chosen to correspond to 15 meV above the $n_z=1$ heavy-hole to conduction band transition. The 20 meV pulse bandwidth produces carriers with kinetic energies <30 meV. Peak carrier densities, as estimated from the pump fluence taking into account multiple reflections within the quantum well stack and mirror layers, were varied in the range $4\times10^{10}-7\times10^{12}\,\mathrm{cm}^{-2}$. The differential reflectivity, which is the pump-induced change in the probe reflectivity expressed as a percentage of the unsaturated probe reflectivity, was measured as a function of probe delay.

Figure 2 shows differential reflectivity data for the samples at a carrier density of $1\times10^{12}\,\mathrm{cm}^{-2}$; also shown is the pulse autocorrelation trace. For ease of comparison, the bleaching signals have been normalized to the peak value near zero delay. In both samples, a rapid increase in absorption saturation is apparent, induced by the pump pulse through state filling. This bleaching signal subsequently decays due to carrier–carrier and carrier-phonon scattering processes which remove carriers from the optically coupled states, leading ultimately to a Fermi distribution of carriers in thermal equilibrium with the lattice. The relatively time-independent plateaus reflect the degree of state filling in the optically coupled states after carriers have thermalized. This ''steady state'' signal decays on a much longer time scale (>50 ps) due to carrier recombination. The most notable difference between the two samples is the much larger steady state bleaching signal, relative to the peak value, in the As-containing sample. After account is taken of the fractional coherent artifact,[9] one finds that the plateau is 19% and 66% of the peak saturation signal for samples with $x=0$ and 0.19, respectively. The decay dynamics also differ significantly in the two samples: exponential fits to the data using a linear prediction singular value decomposition analysis[9–11] indicate a monoexponential decay with time constant $\tau^{x=0.19}=750$ fs in the bleaching trace for the As-containing sample, while data for the GaSb quantum wells exhibits a two-component recovery with time constants $\tau_1^{x=0}=140$ fs and $\tau_2^{x=0}=2.0$ ps [see Fig. 2(b)]. Decay times for GaSb wells were found to be independent of carrier density for densities in the range $2\times10^{11}-7\times10^{12}\,\mathrm{cm}^{-2}$. The single time constant in the As-containing sample was observed to increase from 430 fs at a carrier density of $3\times10^{12}\,\mathrm{cm}^{-2}$–1.8 ps at a density of $4\times10^{11}\,\mathrm{cm}^{-2}$. For lower carrier densities, the decay kinetics in the As-containing sample were observed to be independent of carrier density. (The lowest carrier density employed in the experiments was $4\times10^{10}\,\mathrm{cm}^{-2}$.)

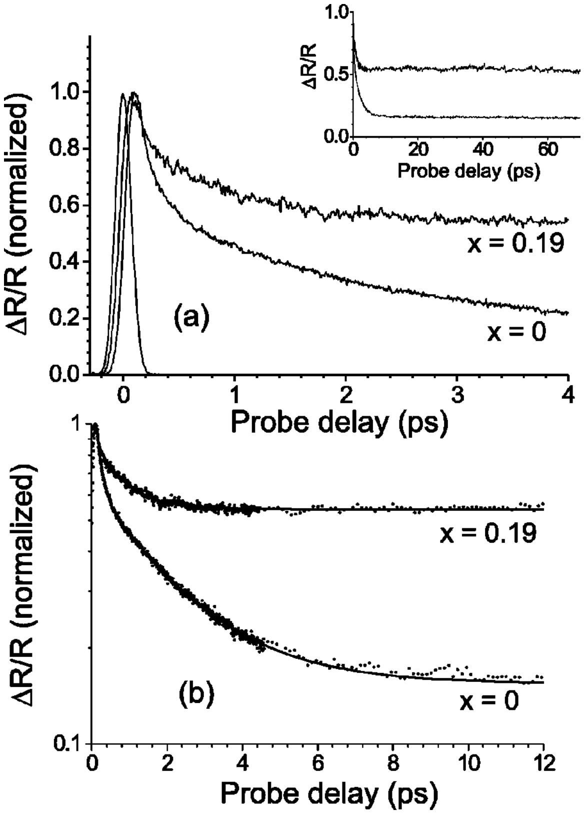

FIG. 2. (a) Data of pump probe reflectivity measurements on $GaAs_xSb_{1-x}/AlSb$ quantum wells for $x=0$ (lower) and $x=0.19$ (upper). Also shown is the pulse autocorrelation. The excited carrier density is $1\times10^{12}\,\mathrm{cm}^{-2}$. Inset: the same data on a longer time scale. (b) Data shown with fitting results, which are indicated by a solid line. A lower resolution of data points was used for $\tau>4.5$ ps for technical convenience.

Since the energy distribution of the injected carriers is the same in the two samples, the differences in differential reflectivity data must reflect differences in the electron scattering processes at the band edge. We attribute the contrasting behavior to the suppression of electron scattering to the L valleys in the As-containing sample due to a larger $L-\Gamma$ energy separation. If injected electrons have access to L valley states, a large fraction of them will rapidly scatter to these valleys via deformation potential scattering. Electron exchange between the Γ and L valleys occurs until equilibrium with respect to both carrier population and thermal energy is attained. In equilibrium, the valley populations are determined by their density of states and the energy separation between the valley edges. Based on the values of E_L-E_Γ from our model (see Fig. 1), we estimate that the percentage of carriers residing in the Γ valley in a Fermi distribution at 295 K would be 10% and 59% in the samples with $x=0$ and 0.19, respectively. Following thermalization, one therefore anticipates a much smaller degree of optically coupled state filling in the GaSb quantum wells than in $GaAs_{0.19}Sb_{0.81}$ wells, and a correspondingly smaller steady-state bleaching signal relative to the peak value, in agreement with the experimental observations.

The difference in decay kinetics also supports this view of the carrier dynamics. In the absence of L valley scattering, injected electrons thermalize to a Fermi distribution with a temperature below 295 K, but approach the lattice temperature through phonon interactions,[12] resulting in a narrow Γ valley energy distribution. The monoexponential decay time of $\tau^{x=0.19} = 750\,\mathrm{fs}$ observed in $\mathrm{GaAs}_{0.19}\mathrm{Sb}_{0.81}$ wells reflects the time taken for thermalization and carrier warming within the Γ valley. The observation of an increase in the recovery time with decreasing carrier density in the data for $\mathrm{GaAs}_{0.19}\mathrm{Sb}_{0.81}$ wells for densities in the range $3\times10^{12}-4\times10^{11}\,\mathrm{cm}^{-2}$ suggests that carrier–carrier scattering plays a role in the carrier dynamics at these densities. For densities below $4\times10^{11}\,\mathrm{cm}^{-2}$, the density-independent recovery time of 1.8 ps is determined primarily by intravalley phonon scattering events.

For the GaSb quantum wells, the observation of a two-component bleaching recovery, with a much faster initial decay ($\tau_1^{x=0} = 140\,\mathrm{fs}$) reflects the presence of the additional $\Gamma-L$ scattering process. Using a deformation potential of $D_{\Gamma-L} = 6\times10^{8}\,\mathrm{eV\,cm}^{-1}$ for GaSb quantum wells, one finds that the $\Gamma-L$ scattering time for electrons resonant with the L valley minima is $\tau_{\Gamma-L} = 70\,\mathrm{fs}$,[4] in reasonable agreement with our results. The longer ($\tau_2^{x=0} = 2\,\mathrm{ps}$) decay time for GaSb wells is determined by carrier thermalization and warming within the Γ valley.

The lack of carrier density dependence in the pump-probe data for GaSb quantum wells indicates that the carrier dynamics during thermalization and warming are determined by phonon scattering processes for the range of densities investigated. The carrier population approaches thermal equilibrium more slowly in GaSb quantum wells than in As-based samples due to carrier exchange with the L valleys: the establishment of equilibrium populations in the Γ and L valleys at a common temperature requires carriers to return from the L valleys. The time constant for L to Γ return scattering using the earlier deformation potential is $\tau_{L-\Gamma} = 2\,\mathrm{ps}$, in agreement with the second measured time constant in the pump-probe data for GaSb quantum wells. The electron return scattering time is much longer than the scatter out time due to the lower density of states in the Γ valley. Electrons returning from the L valleys carry excess energy not only because of the difference in the band offsets, but also because a significant fraction (approximately 40% at 295 K) of the electrons return through phonon absorption. Furthermore, the electrons which scatter to the L valleys warm more rapidly through intravalley phonon scattering than the Γ electrons due to the larger density of states in the L valleys. The returning electrons therefore heat the Γ population,[13] causing the absorption bleaching at the band edge to decrease with time for $\tau < 2$ ps, as the experimental data indicate. Our findings are similar to those reported by Zhou *et al.*,[14] who compared pump-probe results in InP and GaAs using 2 eV, 50 fs pulses.

Our measurements, which were performed with identical initial carrier energy distributions, provide an unambiguous comparison of electron dynamics in structures with and without As. These findings have implications for future optoelectronic applications of Ga(As)Sb quantum wells, where removal of intervalley scattering will improve material characteristics, including internal quantum efficiency and mobility.

The authors gratefully acknowledge financial support of the Natural Sciences and Engineering Research Council of Canada and The Royal Norwegian Research Council, and F. Wise for providing the fitting program.

[1] K. C. Hall, S. W. Leonard, H. M. van Driel, A. R. Kost, E. Selvig, and D. H. Chow, Appl. Phys. Lett. **75**, 4156 (1999).
[2] P. S. Dutta and H. L. Bhat, J. Appl. Phys. **81**, 5821 (1997).
[3] J. Koeth, R. Dietrich, and A. Forchel, Appl. Phys. Lett. **72**, 1638 (1998).
[4] D. C. Smith, E. D. O'Sullivan, L. Rota, A. C. Maciel, and J. F. Ryan, Phys. Status Solidi B **204**, 110 (1997); Physica E **2**, 156 (1998).
[5] *Semiconductors-Basic Data*, edited by O. Madelung (Springer, Berlin, 1996).
[6] In carrying out this calculation, a linear interpolation was used for the relative positions of the Γ and L valleys in the conduction band in the ternary system, which is valid since bowing in the electronic states in Ga(As)Sb is restricted to the valence bands (F. T. Vasko and A. V. Kuznetsov, *Electronic States and Optical Transitions in Semiconductor Heterostructures* (Springer, New York, 1999). Strain effects were included using standard elastic theory [P. Voisin, C. Delalande, G. Bastard, M. Voos, L. L. Chang, A. Segmuller, C. A. Chang, and L. Esaki, Superlattices Microstruct. **1**, 155 (1985)] assuming that the multiple quantum well stack was coherently strained to the Bragg mirror, as indicated by x-ray diffraction and photoluminescence measurements. Confinement was treated using a single band in the envelope function approximation, with an assumed 70% conduction band offset at the Γ point in the heterostructure [F. W. O. Da Silva, C. Raisin, S. Gaillard, C. Alibert, and A. Rocher, Thin Solid Films **190**, 21 (1990)].
[7] The energy of an optical phonon in GaSb is 26 meV (see Ref. 5) and the onset for scattering by phonon absorption occurs at a carrier kinetic energy in the Γ valley that is one phonon energy below the L minimum.
[8] T. C. McGlinn, T. N. Krabach, M. V. Klein, G. Bajor, J. E. Greene, B. Kramer, S. A. Barnett, A. Lastras, and S. Gorbatkin, Phys. Rev. B **33**, 8396 (1986).
[9] A. J. Taylor, D. J. Erskine, and C. L. Tang, J. Opt. Soc. Am. B **2**, 663 (1985).
[10] F. W. Wise, M. J. Rosker, G. L. Millhauser, and C. L. Tang, IEEE J. Quantum Electron. **QE-23**, 1116 (1987).
[11] The analysis was restricted to delay values > than 150 fs, where the pulse autocorrelation falls to <5% of the peak value, to avoid contributions from the coherent artifact in the fitting time constants.
[12] W. W. Rühle and H.-J. Polland, Phys. Rev. B **36**, 1683 (1987).
[13] J. Shah, B. Deveaud, T. C. Damen, W. T. Tsang, A. C. Gossard, and P. Lugli, Phys. Rev. Lett. **59**, 2222 (1987).
[14] X. Q. Zhou, G. C. Cho, U. Lemmer, W. Kütt, K. Wolter, and H. Kurz, Solid-State Electron. **32**, 1591 (1989).