Auger recombination in long-wavelength infrared $\text{InN}_x\text{Sb}_{1-x}$ alloys

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(Received 30 October 2000; accepted for publication 23 January 2001)

Dilute nitrogen alloys of InSb exhibit strong band gap bowing with increasing nitrogen composition, shifting the absorption edge to longer wavelengths. The conduction band dispersion also has an enhanced nonparabolicity, which suppresses Auger recombination. We have measured Auger lifetimes in alloys with 11 and 15 μm absorption edges using a time-resolved pump-probe technique. We find the lifetimes to be longer at room temperature than equivalent band gap $\text{Hg}_{1-y}\text{Cd}_y\text{Te}$ alloys at the same quasi-Fermi level separation. The results are explained using a modified $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian which explicitly includes interactions between the conduction band and a higher lying nitrogen-related resonant band. © 2001 American Institute of Physics. [DOI: 10.1063/1.1355301]

When a small fraction of anion atoms are replaced by nitrogen in GaInAlAs the band gap initially decreases rapidly, by between 0.05 and 0.2 eV at 1% N.¹⁻⁴ This opens the possibility of long wavelength applications in dilute nitride alloys. Despite the decrease in band gap, the conduction band (cb) edge effective mass has been observed to rise with increasing N content,^{5,6} contrary to the prediction of usual $\mathbf{k}\cdot\mathbf{p}$ theory. The strong band gap bowing and increase in effective mass arise because of a repulsive interaction between the conduction band edge and a higher lying band of nitrogen resonant states.⁷⁻⁹

The band structure of $\text{InN}_x\text{Sb}_{1-x}$ can be predicted using a modified $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian previously developed for $\text{GaN}_x\text{As}_{1-x}$.^{7,8} In this model, the Hamiltonian describing the interaction between the cb edge and nitrogen resonant level is given by

$$H_{N,C} = \begin{pmatrix} E_N - \alpha x & \beta \sqrt{x} \\ \beta \sqrt{x} & E_C - \gamma x \end{pmatrix}. \quad (1)$$

The parameters α , β , γ , and the energy of the nitrogen resonant level, E_N , can be found by analyzing the results of a tight binding model we have developed.⁹ In the case of $\text{InN}_x\text{Sb}_{1-x}$, the cb energy $E_C = 0.177$ eV ($\equiv 7 \mu\text{m}$), and we calculate $E_N = 0.647$ eV, $\alpha = \gamma = 0.77$ eV, and $\beta = 2.2$ eV. The band gap is therefore predicted to decrease to 110 meV at 1% N, a fractional change of almost 40%. This clearly offers significant potential for long wavelength emission. III-V materials may be made with band gaps smaller than hitherto possible, opening up a longer wavelength region for direct interband transitions.

Auger recombination (see Fig. 1) is the main recombination scattering process in intrinsic and n -type Kane band

structure materials with band gap less than about 1 eV and, hence, is the primary factor limiting the maximum operating temperature of emitter and detector devices in this energy range. The Auger lifetime varies as

$$1/\tau_{\text{Auger}} = Cn^2, \quad (2)$$

where n is the electron density, and so lasers suffer in particular due to the high values of n at threshold. The Auger coefficient, C , contains a thermal activation energy, E_A , which, in the strongly pumped regime, is given approximately by^{10,11} $E_A = [\mu/(1+\mu)]E_g$. Reducing the band gap,

FIG. 1. Calculated room temperature band structures for $\text{InN}_x\text{Sb}_{1-x}$ alloys with $x=0\%$ (dashed) and 0.6% (solid), for which $E_g=0.177$ and 0.113 eV, respectively. A strong band gap narrowing can be seen as x is increased, without significant reduction in band edge mass. Inset: different types of band structures with the same band gap and with $\Delta F = E_g$. (a) Kane band structure showing a highly degenerate cb, and Auger processes involving highly populated hole states; (b) reduced $m_h \rightarrow m_e$ as in compressively strained layer heterostructures, showing much less degenerate cb, i.e., lower n , than (a) and suppressed Auger processes with higher activation energy; (c) increased $m_e \rightarrow m_h$ as in $\text{InN}_x\text{Sb}_{1-x}$, showing much more degenerate valence band (i.e., higher n) than (a) but, like (b), suppressed Auger.

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E_g , of semiconductor materials with a Kane band structure (to make longer wavelength devices) reduces the effective mass ratio, $\mu = m_e/m_h$ and also E_A .

Considerable experimental and theoretical effort has been devoted to band structure engineering to suppress Auger recombination for long wavelength devices. In compressively strained layer heterostructures a reduced valence band mass has the effect of reducing the excited carrier density required for emission [Fig. 1(b)]. This reduction in n increases τ_{Auger} directly via Eq. (2).¹¹ In addition the increase in μ leads to a reduction in C .

Figure 1 shows the band structure of $\text{InN}_x\text{Sb}_{1-x}$ for $x=0\%$ and 0.6% , calculated using a ten band model (doubly degenerate N level, conduction band, heavy, light, and split-off hole bands). Because of the effects of N incorporation, the room temperature energy gap at $x=0.6\%$ has dropped to 0.113 eV ($\equiv 11\text{ }\mu\text{m}$), but the band edge mass has only dropped from 0.010 to 0.008 . If we set $x=0$ and $E_g=0.113\text{ eV}$ (i.e., considering an equivalent band gap, hypothetical, Kane bandstructure material), the mass drops by twice as much to 0.006 . We note that the only realizable equivalent gap Kane material, $\text{Hg}_{0.83}\text{Cd}_{0.17}\text{Te}$, has a band edge mass yet smaller than 0.006 . In other words the band edge mass is much larger than expected for a Kane material. This may provide a way of reducing C at long wavelengths, even without strain [Fig. 1(c)]. Away from the zone center there is a strong anticrossing where the band meets the N level, and the resulting increased cb nonparabolicity has a similar effect. Finally, the mixing of the localized nitrogen levels into the conduction band (again, more strongly at high k) suppresses the matrix element for Auger recombination. In this work we show experimentally that the Auger lifetime is indeed enhanced over bulk Kane band structure materials, and thus that the N-alloy system is promising for laser applications.

The samples used were undoped and grown by molecular beam epitaxy on semi-insulating GaAs substrates. Sample A had a $3.25\text{ }\mu\text{m}$ InSb strain-relaxing buffer, followed by six repeats of ($33\text{ nm InN}_x\text{Sb}_{1-x}$, 33 nm InSb) nominally with $x=3.5\%$, and $0.5\text{ }\mu\text{m}$ InSb cap. Sample B had a similar buffer and cap, but a $2\text{-}\mu\text{m}$ -thick epilayer of $x=0.4\%$. Sample C was similar to B with $x=0.6\%$.

The time-resolved optical measurements were performed using the Dutch free electron laser (FELIX) and are described in detail elsewhere.¹² A strong pump pulse completely bleaches the transmission by increasing the separation between the quasi-Fermi levels (ΔF) up to the pump photon energy ($\hbar\omega_{\text{pump}}$). The pump-induced transparency was measured with a weak probe pulse delayed in time after the pump. The size of the transmission change is directly related to excited carrier concentration (and the joint density of states at the pump wavelength), but it is not necessary to know this dependence to measure a lifetime. The absorption edge of the sample was determined by measuring the wavelength at which the bleaching effect at zero delay disappears (in effect a modulated transmission technique). The room temperature absorption edges of samples A, B, and C were 11 , 14 , and $15\text{ }\mu\text{m}$, respectively, showing clearly how incorporation of N may move the band gap of III-V alloys to lower energies than previously achieved.

FIG. 2. Differential transmission change as a function of time delay between pump and probe pulses. Symbols: experimental results for sample A ($E_g=0.113\text{ eV} \equiv 11\text{ }\mu\text{m}$) at 290 K , with $\hbar\omega_{\text{pump}}=0.124\text{ eV} (\equiv 10\text{ }\mu\text{m})$. Curves: theoretical transmission change as a function of time all for an initial carrier density $n=4.6\times 10^{17}\text{ cm}^{-3}$. Solid line, $\text{Hg}_{1-y}\text{Cd}_y\text{Te}$ with $y=0.170$ ($E_g=0.113\text{ eV}$ and $\Delta F_{t=0}=0.124\text{ eV}$, i.e., same as our $\text{InN}_x\text{Sb}_{1-x}$ experiment); dashed line, $y=0.216$ ($E_g=0.177\text{ eV}$ and $\Delta F_{t=0}=0.181\text{ eV}$); dotted line, InSb ($E_g=0.177\text{ eV}$ and $\Delta F_{t=0}=0.194\text{ eV}$).

Pump-probe results are shown in Fig. 2 for sample A at room temperature, a pump wavelength of $10\text{ }\mu\text{m}$, and plotted on a log-linear scale so that $1/\tau$ is the slope. There is a rapid decay (steep slope) corresponding to an initial lifetime per excited carrier of 300 ps . The decay rate slows with increasing delay, i.e., with decreasing n , as determined by Auger recombination [Eq. (2)]. After about 200 ps n (and, hence, Cn^2) has reduced sufficiently that the decay has become a single exponential dominated by Shockley-Read-Hall (SRH) processes. The latter process is associated with a constant lifetime (independent of n and t), i.e., a straight line on Fig. 2. This lifetime, which is not an intrinsic property of the material, is quite short, $\tau_{\text{SRH}}=2.6\text{ ns}$, due to the poor morphology of the samples considered. Similar time-dependent lifetime results were obtained at other temperatures and with the other samples—e.g., the initial lifetime for sample B at 10 K (at which temperature the band gap was $0.124\text{ eV} \equiv 10\text{ }\mu\text{m}$) with pump photon energy $0.260\text{ eV} (\equiv 4.9\text{ }\mu\text{m})$ was 130 ps .

We concentrate in the present work on the rate of the Auger process at the moment in time where ΔF is known, i.e., when $\Delta F=\hbar\omega_{\text{pump}}$ immediately after the pump pulse bleaches the system. In order to determine C it would be necessary to know how the transmission varies with n , but this requires detailed knowledge of the density of states, including quantization effects, strain, disorder, etc. We note that the lifetime at the threshold quasi-Fermi level separation is what determines the threshold current in a laser device, and here we determine $\tau(\Delta F)$ unambiguously.

Also shown on Fig. 2 for comparison are the Auger decay curves for Kane band structure materials calculated using the “flat valence band approximation”¹³ (which we have previously shown gives good agreement with experiment in the HgCdTe system¹²). We ignore SRH for this purpose and radiative transitions are negligible in this temperature/density regime. The solid line on the figure is for $\text{Hg}_{1-y}\text{Cd}_y\text{Te}$ with $E_g=0.113\text{ eV} (\equiv 11\text{ }\mu\text{m})$, i.e., the same band gap as sample A, and using a starting ($t=0$) value of $\Delta F=0.124\text{ eV} (\equiv 10\text{ }\mu\text{m})$, i.e., the same as the pump wavelength used in

the experiment. The corresponding initial carrier density is $n = 4.6 \times 10^{17} \text{ cm}^{-3}$. The results show a much shorter initial lifetime of 97 ps. The decay curve for HgCdTe with the same density, but in this case the band gap of InSb ($7 \mu\text{m}$), is also shown, and has an initial Auger lifetime of 270 ps, comparable to that of the N alloy. Finally the decay curve for InSb itself (again with the same density) has an initial lifetime of 500 ps. We can see that the lifetime for the $11 \mu\text{m}$ $\text{InN}_x\text{Sb}_{1-x}$ alloy is only slightly shorter than that for $7 \mu\text{m}$ InSb in spite of its much smaller band gap. However, it is almost as long as that of the $7 \mu\text{m}$ HgCdTe alloy, and much longer than that of the $11 \mu\text{m}$ HgCdTe. These numerical results are not dependent on any prior knowledge of the band structure of the slightly exotic nitride alloy system, its Auger coefficient, or the carrier density, though our conclusion is that the effects observed are a *consequence* of the band structure. Similar suppression over the equivalent band gap HgCdTe was seen for the other alloys, e.g., the HgCdTe equivalent of the 10 K result of sample B is 21 ps (cf. 130 ps mentioned earlier).

In summary, we demonstrate alloys of $\text{InN}_x\text{Sb}_{1-x}$ with transmission edge wavelengths out to $15 \mu\text{m}$ at room temperature. We show that for a given quasi-Fermi level separation the Auger recombination rate is about one third that of equivalent band gap HgCdTe, due to the higher electron mass and conduction band nonparabolicity. As a result of this, all other things being equal, a $10 \mu\text{m}$ wavelength laser based on $\text{InN}_x\text{Sb}_{1-x}$ material would have an Auger current at threshold less than an equivalent HgCdTe device, highlight-

ing the potential of this material for optoelectronic applications.

The authors are grateful to EPSRC (UK) for support. The work was carried out as part of the EPSRC (UK) and FOM (NL) experimental program at FELIX and the authors are especially grateful for the skillful assistance of Dr. A. F. G van der Meer. The authors would also like to thank Dr. M. White for the use of his Auger calculation spreadsheet.

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