Band anticrossing in dilute $\text{InN}_x\text{Sb}_{1-x}$

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(Received 20 December 2001; accepted for publication 17 May 2002)

Dilute nitrogen alloys of InSb exhibit extremely strong band gap bowing with nitrogen composition that has been associated with anticrossing between the localized resonant states of the nitrogen within the conduction band and the extended states of the conduction band itself. This also results in the conduction band dispersion having an enhanced nonparabolicity. We have measured the electron effective mass near the anticrossing by cyclotron resonance in $\text{InN}_x\text{Sb}_{1-x}$ alloys with absorption edge near $15\ \mu\text{m}$, using pulsed fields up to 150 T. The results directly demonstrate the band anticrossing and quantitatively confirm the increase of effective mass versus x predicted for $\text{InN}_x\text{Sb}_{1-x}$ by a tight binding calculation for low nitrogen concentration ($x < 0.01$). © 2002 American Institute of Physics. [DOI: 10.1063/1.1493663]

It is by now well known that partial replacement of group V elements with dilute nitrogen atoms in AlGaAs produces a very strong downward bowing of the band gap to longer wavelengths than the host, with a shift between 0.1 to 0.2 eV at 1% N.^{1–3} This has opened up the possibility of long wavelength emission even in very dilute alloys. Despite the decrease in band gap, the conduction band effective mass has been predicted to rise with increasing N content, contrary to the expectation of usual $\mathbf{k}\cdot\mathbf{p}$ theory.^{4,5} The strong band gap bowing and increase in effective mass have been explained by a band anticrossing model in which resonant localized N states within the conduction band interact with the extended states of the conduction band.^{6–9} The effective mass increase has been determined by the plasma reflection technique and shown to be consistent with pressure dependent differential reflectivity measurements.^{4,5}

This dramatic band gap bowing has also been observed in alloys of narrow gap semiconductors and we have previously measured both this, and the resulting suppression of Auger recombination in $\text{InN}_x\text{Sb}_{1-x}$ using a time resolved pump–probe technique. We showed¹⁰ that for a given quasi-Fermi level separation the Auger recombination rate is about one third that of the equivalent band gap HgCdTe (band gap between 10 and $15\ \mu\text{m}$), and proposed that this is due to the higher conduction band nonparabolicity.¹¹ We here demonstrate by direct measurement of cyclotron resonance (CR) at high pulsed magnetic fields that the electron effective mass of $\text{InN}_x\text{Sb}_{1-x}$ (with $0 < x < 1\%$) is substantially higher than that of InSb, and directly measure the band anticrossing effect. Furthermore, we find that the increase in effective mass with x value is in excellent quantitative agreement with a

tight-binding calculation of the interaction coefficients.⁸

The band structure of $\text{InN}_x\text{Sb}_{1-x}$ with small x can be predicted using a modified $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian previously developed for $\text{GaN}_x\text{As}_{1-x}$.^{6,8} In this model, the energy of the nitrogen resonant level, E_N and of the host conduction band (cb) edge energy, E_C , both shift linearly with increasing N composition, x , while the repulsive interaction between resonant states and the cb edge increases as $x^{1/2}$. There are, in addition, x - and k -dependent terms for the interaction between the nitrogen and valence bands, but these have been shown to be small even far from the zone center and we neglect them here.

Incorporating a magnetic field in this approximation is trivial, as the lack of k -dependent terms means there are no additional raising and lowering operators, and hence no additional warping terms which would couple Landau levels with a different quantum number. In our case, because of the narrow band gap of the host, even before the addition of the nitrogen, there is a strong $\mathbf{k}\cdot\mathbf{p}$ interaction between the conduction and valence bands, so we use the 8×8 Pidgeon and Brown (PB) model¹² as the basis:

$$H = \begin{bmatrix} E_N + \alpha x & \beta\sqrt{x} & 0 & 0 & 0 \\ \beta\sqrt{x} & H_{CC} + \gamma x & H_{CH} & H_{CL} & H_{CS} \\ 0 & H_{HC} & H_{HH} & H_{HL} & H_{HS} \\ 0 & H_{LC} & H_{LH} & H_{LL} & H_{LS} \\ 0 & H_{SC} & H_{SH} & H_{SL} & H_{SS} \end{bmatrix}, \quad (1)$$

where H_{ij} are the PB matrix elements between cb (C), light (L), heavy (H), and spin–orbit split off (S) valence bands. Following PB, we assume $k_z = 0$ and decoupling the spin-up and spin-down terms of the resulting 10×10 Hamiltonian, we have two 5×5 eigenproblems to solve. The band gap, spin–orbit split-off band gap, conduction–valence interac-

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FIG. 1. Calculated room-temperature Landau level spectrum ($n=0$ to 3) vs applied magnetic field for (a) InSb, i.e., $x=0$, showing the virtual N level at $E_N=0.6$ eV above the cb edge, and (b) $\text{InN}_x\text{Sb}_{1-x}$ with $x=0.8\%$. Vertical bars show $n=0$ to 1 CR (i.e., spin conserving) transitions for $10.6\ \mu\text{m}$ and $5.5\ \mu\text{m}$.

tion parameter and Luttinger valence band parameters for the InSb binary were taken from Ref. 13. The parameters α , β , γ , and E_N can be found by analyzing the results of a tight-binding model.⁸ In the case of $\text{InN}_x\text{Sb}_{1-x}$, $E_G=0.177$ eV ($\approx 7\ \mu\text{m}$) at room temperature, and we calculate the nitrogen level energy, $E_N=0.47$ eV above the cb edge, $\alpha=-2.77$ eV, $\beta=2.2$ eV, and $\gamma=0.0$ eV.^{8,10}

An example of the cb Landau level spectrum produced by this model with the aforementioned parameters, is shown in Fig. 1 for InSb (i.e., with $x=0$) and for an alloy with $x=0.8\%$. The first effect of increasing x is that the N-like band rises and the Γ -like cb drops in energy. The band gap is predicted to decrease by about 80 meV per percent N, a fractional change of 40%, as shown in Fig. 2. This clearly offers significant potential for long wavelength emission/detection from direct band gap III-V materials. At the same time as the band gap is reduced, the effect of mixing the localized nitrogen states with the Γ -like cb causes a very strong anticrossing and tends to *increase* the effective mass.

FIG. 2. Calculated band gap energy for $\text{InN}_x\text{Sb}_{1-x}$ alloys vs nitrogen composition x .

FIG. 3. CR of the $\text{InN}_x\text{Sb}_{1-x}$ alloy at room temperature using left circular polarized light.

Very close to the zone center (at energies less than a few tens of meV and fields below about 10 T), the $\mathbf{k}\cdot\mathbf{p}$ repulsion of the valence band is stronger and the band edge mass decreases slightly, although this is only just discernable on the scale in Fig. 1. A few tens of meV above the cb edge, the nitride has a heavier mass than the binary. The only Kane bandstructure materials without nitrogen but with an equivalent band gap, $\text{Hg}_x\text{Cd}_{1-x}\text{Te}$, have a much smaller mass than either the binary or the nitride.

We have performed pulsed field CR measurements on a $2.4\ \mu\text{m}$ thick epilayer of $\text{InN}_x\text{Sb}_{1-x}$ nominally with $x=1.1\%$. The sample was grown by molecular-beam epitaxy on a semi-insulating GaAs substrate and had a $0.4\ \mu\text{m}$ buffer layer/virtual substrate of intrinsic InSb between the GaAs and the nitrided layer. The room-temperature n -type carrier concentration resulting from nitrogen incorporation was $3.87\times 10^{17}\ \text{cm}^{-3}$. The carrier mobility was of $\sim 5500\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, necessitating a high magnetic field and short wavelength to achieve the necessary CR observation condition, $\omega_c\tau \gg 1$. We have used the single turn coil technique at the University of Tokyo which generates high magnetic fields with a very short pulse duration of $6\ \mu\text{s}$.¹⁴ The radiation sources for the high-field results were CO_2 (9.2 and $10.6\ \mu\text{m}$) and CO ($5.5\ \mu\text{m}$) lasers.

Magnetoabsorption results are shown in Fig. 3 at room temperature for three different exciting laser wavelengths. The structure observed near 10 T for $5.53\ \mu\text{m}$ is the interband spin doublet from the undoped InSb buffer layer. The other peaks are the spin-split CR of the n -type epilayer. The interband structure is not observed for the nitride epilayer due to the doping, which gives a Fermi level well above the band edge at a low field. Similarly, the CR is not observed for the intrinsic InSb buffer as the total areal density of carriers is two orders of magnitude less than for the nitride layer.

The CR energies are plotted as a function of magnetic field strength, together with the theoretically computed energies for the aforementioned model in Fig. 4. The theoretical lines have been fitted to the data by varying only two of the available parameters, x and E_N . None of the InSb host parameters were treated as adjustable, and are taken from Ref. 13, along with the room-temperature band gap of 0.177 meV. Using the InSb parameters, the two lowest left circularly polarized (i.e., light hole) interband transitions are at 7.4 T

FIG. 4. CR transition energy vs magnetic field. Points: peak positions from data of Fig. 3, solid line: fitted spin-up and spin-down CR positions using the data from Fig. 1, and dashed line: spin-up and spin-down CR positions for InSb, also from Fig. 1.

and 13.6 T, with optical matrix elements in the ratio 3:1, i.e., correspond exactly to the low-field features at $5.53 \mu\text{m}$. Similar agreement is seen for the right circular polarized heavy hole transitions (data not shown).

The calculation of the nitride CR energies is quite insensitive to the parameters α and γ . Because of this, varying x and β have nearly the same effect [see Eq. (1)], and because of the uncertainty in the fraction of nitrogen atoms which end up substituting with Sb, we used x as a fitting parameter. The calculation of Fig. 4 used $x=0.8\%$, and $E_N=0.57$ eV above the cb edge.

At the highest photon energy, the spin-down line has disappeared, or became weak and very broad (there is possibly a vestigial feature at 130 T in Fig. 3). This is due to the anticrossing with the nitrogen levels, which is reached first by the higher-energy spin-down transition. It should return to extended Γ -like character and reappear at higher fields. The spin-up line also anticrosses, at about 180 T.

An alternative explanation for the disappearance would be that the quantum limit has been achieved, and the carriers have all relaxed to the lowest spin state. We discount this as unlikely, since it has been shown in identical pulsed field CR experiments on InAs quantum wells that both peaks of the spin doublet are normally seen on the up sweep, due to a very slow spin relaxation time in comparison with the duration of the field pulse.¹⁵

The effective mass of the $5.53 \mu\text{m}$ transition is 0.044, as compared with 0.033 calculated for InSb at that energy. We note that it is the effective mass at energies comparable with E_g above the cb edge which is important in Auger recombination. The heavier this mass, the slower the Auger rate, and this is consistent with our earlier measurement¹⁰ of increased recombination times in these materials.

In summary, we have performed very high-field magneto-optical absorption measurements on $\text{InN}_x\text{Sb}_{1-x}$, and have directly shown the influence of the nitrogen resonant states on increasing the electron effective mass.

The authors are grateful to EPSRC (UK) for support and to E. O'Reilly, A. Andreev, and N. James for helpful discussions on theory.

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