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Direct evidence for suppression of Auger recombination in GaInAsSbP/InAs mid-infrared light-emitting diodes

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Mid-infrared light emitting diodes based on the pentanary alloy GaInAsSbP have been engineered to provide a favourable band structure for the suppression of non-radiative Auger recombination which is dominant in narrow band gap III-V materials. Hydrostatic pressure measurements at room temperature and at 100 K were used to tune the band gap towards resonance with the spin-orbit band gap. Analysis of the resulting electroluminescence confirms that the non-radiative Auger recombination process involving the spin-orbit-split-off-band is suppressed under ambient conditions. © 2011 American Institute of Physics. [doi:10.1063/1.3646910]

Mid-infrared (2–5 μm) light-emitting diodes (LEDs) are of increasing interest for a variety of technologically important applications including environmental gas monitoring, industrial process control, non-invasive medical diagnosis, and infrared spectroscopy.¹ However, the narrow band gap and density of states imbalance in InAs and related alloys give rise to strong non-radiative Auger recombination and inter-valence band absorption, which makes uncooled room temperature operation difficult to achieve.² LEDs emitting within this spectral range have previously been produced based on bulk InAsSb (Ref. 3) or AlInSb.⁴ LEDs with active regions containing InAsSb/InAs quantum wells,⁵ InSb quantum dots,⁶ and InAs/GaSb superlattices⁷ have also been investigated.

An attractive alternative approach is to use the pentanary alloy GaInAsSbP, because the presence of the fifth element allows an additional degree of freedom for tailoring the band structure, while keeping the lattice parameter constant.⁸ Two of the primary Auger recombination mechanisms in narrow band gap semiconductors are recombination of a conduction band electron with a heavy-hole which produces a hole in the spin-orbit-split-off-band (known as conduction band to heavy hole band and spin-orbit split-off band to heavy hole band (CHSH) recombination) and conduction electron to heavy-hole where another conduction electron is excited to higher energy (known as conduction band to heavy hole band and conduction band to conduction band (CHCC) recombination). These are known to be strongly temperature dependent with a cubic dependence on the carrier concentration. Increasing the spin-orbit split-off energy (Δ_{so}) to be larger than the bandgap (E_g) should help suppress CHSH non-radiative Auger recombination which becomes increasingly important near resonance when $E_g \sim \Delta_{\text{so}}$. Previously, we demonstrated room temperature GaInAsSbP pentanary LEDs operating in the 3–4 μm spectral range.^{8,9} In this work, we report on the behaviour of the electroluminescence (EL) under the application of high hydrostatic pressure to investigate directly the effect on CHSH Auger suppression.

Undoped epitaxial layers of pentanary $\text{Ga}_{0.03}\text{In}_{0.97}\text{As}_{0.83}\text{Sb}_{0.14}\text{P}_{0.03}$ ($\sim 2 \mu\text{m}$ in thickness, $E_{g(300\text{ K})} \sim 0.32 \text{ eV}$) were grown nominally lattice-matched onto n-type ($2 \times 10^{18} \text{ cm}^{-3}$) InAs substrates (100) by liquid phase epitaxy using a horizontal sliding boat in the ultrapure H_2 atmosphere. A wider band gap ($E_{g(300\text{ K})} \sim 0.44 \text{ eV}$) $\text{p}^+ \text{InAs}_{0.62}\text{Sb}_{0.14}\text{P}_{0.24}$ quaternary layer ($\sim 1 \mu\text{m}$ in thickness) was then grown to form the p - n heterojunction. The compositions were confirmed by energy-dispersive x-ray spectroscopy (EDX), and the mismatch was found to be $<0.3\%$ from x-ray diffraction measurements. Details of the growth procedure have been previously reported.^{9,10} Circular mesa diodes were subsequently fabricated with diameters of 300 μm and 750 μm using conventional photolithography to form the LEDs.

High hydrostatic pressure measurements on semiconductor devices have the benefit of being non-destructive and reversible and can yield valuable information on recombination mechanisms.¹¹ Such pressure measurements were performed over the range of 0–8 kbar using compressed helium gas from a U11 gas compressor and a GOC102 optical cell with a sapphire window. Low temperature measurements were performed with the aid of a closed cycle cryostat. The device was pulsed using an Agilent 8114 A pulse generator (pulse width of 500 ns at a frequency of 10 kHz) to minimise Joule heating. EL was detected using a cooled (77 K) InSb detector and an EG&G 7265 lock-in amplifier. High pressure spectral measurements were performed using a Fourier transform infrared (FTIR) system.

Figure 1 shows the EL spectra of the 750 μm diameter device at room temperature and 100 K measured under quasi-cw operation (50% duty cycle) to maximise the signal intensity at a current density of 23 A/cm². The 300 K emission was found to peak at 3.73 μm (0.332 eV), consistent with a band gap of 0.319 eV after subtracting $kT/2$ to account for the thermal carrier distribution. The emission spectra at atmospheric pressure over a wide range of temperatures have been previously reported.⁹ No evidence of radiative recombination was observed from the substrate.

For mid-infrared LEDs, the total current, I , may be written in terms of Eq. (1), where e is the electron charge, V is the active region volume, and A , B , and C are the Shockley-

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FIG. 1. (Color online) Forward-bias normalized EL spectra at 100 K and room temperature. Quasi-continuous operation at 10 kHz with a 120 mA drive current.

Read-Hall (SRH), radiative, and Auger coefficients, respectively. I_{leak} is the leakage current and is thought to be pressure insensitive (inter-valley scattering is negligible due to the large Γ -L and Γ -X splitting values. Direct hetero-junction leakage is believed to be negligible due to pressure independent current voltage characteristics. As all materials used in the device are InAs-rich, the band offsets were assumed to be independent of pressure because the pressure coefficients (dE/dP) of the band gaps of the active region and barriers are approximately the same. SRH recombination is also almost independent of pressure due to the capture cross section for recombination through impurities being pressure insensitive,²¹

$$I = eV(An + Bn^2 + Cn^3) + I_{leak}. \quad (1)$$

An expression for the Auger coefficient is given in Eq. (2), where C_0 is approximately independent of pressure and temperature, and E_a is the activation energy of the Auger process. Assuming parabolic and isotropic bands, the activation energies for CHSH and CHCC recombinations are given by Eqs. (3) and (4), where E_g is the direct band gap energy and m_{so} , m_h , and m_e are the spin-orbit, heavy-hole, and electron effective masses, respectively,¹²

$$C \approx C_{0(CHSH)} \exp(-E_a/kT) + C_{0(CHCC)} \exp(-E_a/kT), \quad (2)$$

$$E_a(CHSH) = m_{so}(E_g - \Delta_{so}) / (2m_h + m_e - m_{so}), \quad (3)$$

$$E_a(CHCC) = m_e E_g / (m_e + m_h). \quad (4)$$

Under the condition that $E_g = \Delta_{so}$, CHSH Auger recombination becomes resonant, as evident from Eq. (3) since $E_a \rightarrow 0$. In practical terms, if $\Delta_{so} > E_g$, then the CHSH process will be suppressed.¹³ Systematic photoreflectance measurements on GaInAsSbP (Refs. 14 and 15) have revealed that $\Delta_{so} = 0.455 \pm 0.010$ eV for the pentanary alloy used in the LEDs and that this value is approximately independent of temperature. Hence, since $E_g = 0.319$ eV, CHSH Auger recombination is expected to be negligible under ambient conditions in these LEDs.

FIG. 2. (Color online) Band gap energy shift with increasing pressure at 100 K in 10 kHz pulsed operation with a drive current of 100 mA. Also shown is the expected pressure dependence at 300 K, assuming dE/dP is temperature insensitive as experimentally confirmed in other III-V devices.

The change in band gap with respect to hydrostatic pressure, dE/dP , has been well characterised for III-V binary compounds and various ternary alloys.¹⁶ However, work on quaternary and pentanary compounds is somewhat less abundant. In order to determine the band gap dependence on pressure for the pentanary alloy, we used linear interpolation between GaAsSbP and InAsSbP quaternary alloys.¹⁷ It was calculated that $dE/dP = 10.6$ meV/kbar at room temperature, which is close to that of InAs at 10.0 meV/kbar.¹⁸ EL spectral data were obtained as a function of pressure from which the emission peak energies at 100 K are plotted as shown in Figure 2. A distinct linear relationship was found, corresponding to a dE/dP value of 10.7 ± 0.3 meV/kbar, in excellent agreement with the theoretical value. This value is expected to be insensitive to temperature as observed for other III-V alloys.^{19,20}

Figure 3 shows the integrated spontaneous emission as a function of pressure at 290 K and 100 K. Since the application of pressure causes an increase in the band gap while maintaining an approximately constant Δ_{so} , it can be used to investigate the behaviour of the Auger processes without changing the device temperature. The spontaneous emission rate increases monotonically with applied pressure, whereas for the CHCC process, increasing pressure causes a decrease in the Auger coefficient, and hence, the EL output increases. For the CHSH process, if Δ_{so} is initially larger than E_g , as here, pressure can bring the CHSH process into resonance as E_g increases towards Δ_{so} , therefore, causing the EL output to decrease. Thus, by measuring the output emission intensity of GaInAsSbP LEDs at a fixed current versus pressure, one may observe the competing effects of these processes as shown in Fig. 3. Two devices were tested, with the same structure and nominally the same composition. Device 1 had an emitting area $750 \mu\text{m}$ in diameter, while device 2 had a smaller diameter of $300 \mu\text{m}$. The measured integrated EL emission (proportional to the radiative current flowing through the device) increased by a factor of 1.75 from atmospheric pressure to 6 kbar (device 1, 290 K). The initial increase in radiative output with applied pressure was smaller than that of previous type II InAs LEDs emitting at $3.3 \mu\text{m}$,²¹ which exhibited a $4 \times$ increase from 0-10 kbar, indicating that Auger recombination is less of a problem in the pentanary GaInAsSbP LEDs. At 290 K, an increase in

FIG. 3. (Color online) Light-pressure curves of two GaInAsSbP LEDs at 100 K and 290 K using 500 ns pulses at 10 kHz with a drive current of 150 mA. The lines are fit as a guide to the eye.

integrated emission is observed with increasing pressure with a decrease in gradient at higher pressure tending towards a maximum at ~ 7.4 kbar. At 100 K, a stronger increase in intensity is observed, which reaches a maximum at ~ 3.6 kbar at low temperature. We calculated that only 25% of the initial increase is due to the increase in the radiative recombination coefficient with pressure.²² The majority of the increase in EL originates from a reduction in Auger recombination with increasing pressure, due to the increase in the activation energy of the CHCC Auger process as the band gap increases.

The existence of the intensity maximum is consistent with the onset of the CHSH process as the band gap becomes equal to the spin-orbit splitting. The difference in pressure at the maxima (7.4–3.6 kbar) corresponds to a difference in band gap change of ≈ 41 meV (using $dE/dP = 10.7$ meV/kbar). Previously reported temperature dependence measurements⁹ provide a value of dE_g/dT at temperatures above 100 K of ≈ 0.24 meV/K. Hence, the band gap increases by 45 meV with cooling down from 290 K to 100 K. This value is approximately equal to the observed difference in the band gap energies at the intensity maxima as shown in Figure 3. At room temperature, with increasing pressure, the intensity initially increases due to the decrease in the CHCC Auger process. However, as E_g approaches Δ_{so} the CHSH process becomes significant causing the intensity to decrease. The two processes together (CHCC decreasing and CHSH increasing with increasing pressure) give rise to the observed maximum. At 100 K, the band gap is initially ~ 45 meV larger, and hence, E_g approaches Δ_{so} at a lower pressure (since, Δ_{so} is relatively temperature insensitive¹⁹). This provides strong evidence that the CHSH Auger process has been suppressed at atmospheric pressure. A similar phenomenon has previously been observed in GaInAsSb lasers operating at $2.37 \mu\text{m}$ using high pressure techniques.²³ Since the spin-orbit split-off energy is largely controlled by the group V composition, it is relatively insensitive to pressure. Hydrostatic pressure also increases both m_{so} and m_c .²⁴ From the low temperature intensity dependence on pressure and using the relation shown in Figure 2, Δ_{so} is

expected to be greater than 0.44 eV, compared to a band gap of 0.37 eV, thus confirming the CHSH resonance detuning in the pentanary alloy. This value is in good agreement with the value of 0.455 ± 0.010 eV obtained using spectroscopic techniques.¹⁴

In summary, we have shown the first direct experimental evidence of CHSH suppression in pentanary LEDs using high pressure and low temperature measurements. Our findings highlight the possible advantages of GaInAsSbP alloys for band-structure engineering of higher efficiency mid-infrared emitters.

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