High-pressure measurements of mid-infrared electroluminescence from InAs light-emitting diodes at 3.3 μm

S. A. Choulis, A. Andreev, M. Merrick, A. R. Adams, and B. N. Murdin

Department of Physics, Advanced Technology Institute, University of Surrey, Guildford, GU2 7RX, United Kingdom

A. Krier^{a)} and V. V. Sherstnev

Department of Physics, Lancaster University, Lancaster, LA1 4YB, United Kingdom

(Received 14 November 2002; accepted 2 January 2003)

The spontaneous electroluminescence emission of InAs light-emitting diodes (LEDs) operating at 3.3 μm was studied as a function of applied hydrostatic pressure. An enhancement of a factor of almost four in radiative efficiency at room temperature was observed in the range 0 to 10 kbar. Analysis of the dependence of electroluminescence emission intensity on hydrostatic pressure at constant current reveals that nonradiative Auger recombination dominates the quantum efficiency of these LEDs. © 2003 American Institute of Physics. [DOI: 10.1063/1.1555276]

InAs-based light-emitting diodes (LEDs) operating in the 2–5 μm spectral region have great potential as the basis of key components in the next generation of optical gas sensors.¹ However, their external quantum efficiency at room temperature is low ($<0.5\%$) and consequently, new mid-infrared analytical and sensor techniques remain predominantly laboratory bound. The principal challenges for efficient room-temperature operation are to reduce nonradiative Shockley–Read and Auger recombination processes, which spoil the internal efficiency, and then to improve optical out-coupling of the generated light.

The application of hydrostatic pressure has been extensively used as an investigative tool to study the internal efficiency and recombination mechanisms that occur in optoelectronic devices.² The major advantage of high-pressure investigations is that they enable nondestructive and reversible variation of the semiconductor band gap, which yields information about radiative and nonradiative recombination rates. We note that although hydrostatic pressure has been extensively used to study semiconductor LEDs and laser structures, the performance of mid-infrared LEDs subjected to high pressure has not previously been investigated. In this letter, we report on the influence of applied hydrostatic pressure (up to 10 kbar) on 3.3 μm mid-infrared InAs LEDs operating at room temperature, using He gas as the pressure medium.

The 3.3 μm LEDs for this investigation were grown on InAs substrates by liquid-phase epitaxy (LPE), using a conventional horizontal sliding boat in ultrapure hydrogen. The devices were of a double-heterostructure design in which the unintentionally doped n -InAs active layer was enclosed between p - and n -InAsSbP confinement layers. The P content in the confinement layer was 0.40 ($E_g = 570$ meV) to provide a higher band-gap energy and large interface band offsets for good carrier confinement. The InAs active region was 0.7 μm thick ($E_g = 354$ meV, under ambient conditions). The resulting samples were processed into mesa-

etched LEDs using standard photolithographic techniques. Fabrication details and device characteristics when operating under ambient conditions have been published previously elsewhere.³

The InAs/InAsSbP double heterostructure exhibits type-II behavior and although it does not contain any quantum wells, band bending at the heterostructure interface leads to some confinement of the carriers in electron–hole quasi-quantum wells located on either side of the interface. This affords the possibility of reduced Auger recombination under ambient conditions since the electron–hole wavefunction overlap is reduced. The application of hydrostatic pressure can give information on the relative significance of the different recombination mechanisms that are sensitive to changes in the band gap, as shown below. Under ambient conditions ($T = 298$ K, $P = 1$ atm), the LEDs exhibited electroluminescence with a peak wavelength of 3.3 μm .³ Light-current characteristics were measured for several LEDs as a function of hydrostatic pressure using 10 μs current pulses of up to 140 mA at a frequency of 5 kHz (corresponding to a duty cycle of 5%). After passing through the pressure cell window, the radiation was collected using a cooled (77 K) InSb detector and a lock-in amplifier. Note that the emission was viewed through the n -type (top) side of the LED, which was 300 μm in diameter and had a ring-like contact. This allows us to neglect intervalence band absorption in the following interpretation, as this process only occurs in p -type layers. Figure 1 shows a plot of the integrated spontaneous electroluminescence (L_{spont}), corrected for detector response, against current in the pressure range 0 to 9.4 kbar for one of the 3.3- μm LED samples. We observed a significant increase in radiative efficiency as a function of hydrostatic pressure. Figure 2 shows that for two different devices, the light output at constant current (140 mA) increased by a factor of 3.74 over the range from 0 to 9.4 kbar. We can give a qualitative interpretation of this in terms of the relative contribution of the radiative and Auger recombination mechanisms in our LEDs. The current density I through the device can be expressed as

^{a)}Electronic mail: a.krier@lancaster.ac.uk

FIG. 1. Measured integrated spontaneous emission intensity as a function of current for $0 \text{ kbar} \leq P \leq 9.4 \text{ kbar}$ at room temperature for one of the InAs/InAsSbP $3.3 \mu\text{m}$ LEDs.

$$I = I_{\text{leak}} + e d_{\text{eff}} (A n + B n^2 + C n^3), \quad (1)$$

where I_{leak} is the leakage current, e is the electron charge, d_{eff} is the effective width of the active region where the radiative recombination occurs, n is the carrier density, and A , B , and C are the usual recombination coefficients for Shockley–Read–Hall (SRH), radiative, and Auger recombination, respectively. Due to device geometry (ring contact with relatively large contact area), we assume that the I_{leak} term is mainly due to electron and/or hole leakage over the heterojunction barriers. However due to the large InAsSbP/InAs band offsets and due to the fact that no light emission was observed corresponding to the bulk band gaps of InAs and InAsSbP³ we believe that I_{leak} is insignificant under ambient conditions. Furthermore, since the bands are expected to behave similarly in these materials under applied pressure, the leakage may be considered as constant. For example, we estimate that the corrections to the electron and hole band offsets induced by a pressure of 9.4 kbar are only 4 and 1 meV, respectively. We therefore assume that I_{leak} is independent of pressure. Furthermore, the devices studied here were produced using a rare-earth gettering technique,⁴ which re-

duces the residual doping in the active region to $< 5 \times 10^{15}$ and hence we may regard the associated SRH recombination [An term in Eq. (1)] as insignificant at room temperature (previous experiments also support this conclusion^{1,3}). In any case, the capture cross section for recombination through defects or impurities is almost independent of pressure. Therefore, it is clear from the large change in electroluminescence with applied pressure which we observed, that neither leakage nor SRH recombination can be the dominant mechanism in these devices. We may assume that at constant current the sum of the radiative and Auger terms in Eq. (1) is pressure independent:

$$e d_{\text{eff}} (B n^2 + C n^3) = \text{const}(P). \quad (2)$$

The measured integrated spontaneous emission is proportional to $L \propto B n^2$ and increases by a factor of 3.74 when P is increased from 0 to 9.4 kbar. From Eq. (2) we estimate that the contribution of radiative recombination $I_{\text{rad}} = e d_{\text{eff}} B n^2$ to the current density at zero pressure is less than 27%, which means that Auger recombination is the dominant process in these LEDs.

Type-II heterostructures have two important channels for Auger recombination; conduction–heavy-hole-band to conduction–conduction-band (CHCC) and conduction–heavy-hole to heavy-hole–split-off-band (CHHS).⁵ Simple analytical expressions can be used to understand how these processes vary with band gap. The band-to-band Auger process can be expressed as a thermally activated process given by⁶

$$C = C^0 \exp\left(\frac{-\Delta E_{\alpha}}{\kappa T}\right) \Leftrightarrow \ln C = \ln C^0 - \frac{\Delta E_{\alpha}}{\kappa T}, \quad (3)$$

where C^0 is approximately independent of temperature and pressure, but includes terms due to the Coulomb interaction between initial and final Auger states. The main pressure dependence of the Auger activation energy (ΔE_{α}) for the direct Auger recombination processes CHCC and CHHS is due to the band-gap dependence of ΔE_{α} and is given by⁶

$$\begin{aligned} \Delta E_{\alpha}(\text{CHCC}) &= \frac{m_e E_g}{(m_e + m_h)}, \\ \Delta E_{\alpha}(\text{CHHS}) &= \frac{m_{\text{so}}(E_g - \Delta_{\text{so}})}{(2m_h + m_e - m_{\text{so}})}, \end{aligned} \quad (4)$$

where m_e ($0.023m_0$), m_h ($0.41m_0$), and m_{so} ($0.16m_0$) are the electron, hole, and spin–orbit effective mass, respectively (the values within the parentheses are the effective masses of InAs under ambient conditions used in the calculations). Δ_{so} is the spin–orbit split-off energy. The electron and spin–orbit effective masses increase approximately proportionally with increasing pressure, while the hole effective mass is relatively independent of pressure.² From Eqs. (3) and (4), it can be seen that the Auger coefficient significantly reduces with increasing E_g and hence with increasing pressure ($\partial E_g / \partial P$ for InAs is 10 meV/kbar ⁷).

Although in the InAs active region, the band gap and the spin–orbit band gap are nearly equal, experimentally we do not observe any resonant behavior in the light output as a function of applied hydrostatic pressure, as suggested by Eq.

FIG. 2. Electroluminescence emission intensity (L_{spon}) at constant current, $I = 140 \text{ mA}$, measured as a function of pressure for two of the InAs/InAsSbP LEDs studied.

FIG. 3. A comparison of the pressure dependence of the electroluminescence emission intensity for type-I (3.8 μm) and type-II (3.3 μm) double-heterostructure LEDs at 300 K.

(4) (see Fig. 1). Consequently, we believe that over the full pressure range, the CHCC process is more dominant.

We performed a similar experiment on double-heterostructure type-I LEDs, which were also fabricated in our laboratory by LPE.³ These LEDs had an $\text{InAs}_{0.95}\text{Sb}_{0.05}$ active region and operated at 3.8 μm at room temperature. The results are compared in Fig. 3 for a constant injection current of 140 mA in both cases. There is clearly a much larger increase in electroluminescence from the type-II LED compared with the type-I LED. The 3.7 increase in the radiative efficiency of the type-II LED compared to 1.7 in the case of the type-I is experimental indication that Auger processes are more important for the type-II LED compared to the type-I LED. However, it is well known that a type-II structure reduces the overlap between electron and hole wavefunction since the carriers are delocalized. This should result in a reduction of the Auger coefficient C in type-II structures compared to type I. However, our results indicate stronger Auger suppression with applied pressure for type-II rather than type-I structures. However, the effective width of the recombination zone within the active region of the type-II LED, (d_{eff}) is significantly smaller. Because of type-II band edge alignment, the injected carriers become localized near the interface, and the electrons and holes must tunnel through potential barriers to the left/right of the confinement regions before recombining with each other. Therefore, the value of d_{eff} is essentially determined by these tun-

neling distances, and we estimate a value of $d_{\text{eff}} \sim 150 \text{ \AA}$. We suppose that, although type-II recombination across the $\text{InAs}/\text{InAsSbP}$ interface reduced the Auger coefficient C , the injected carrier concentration is actually greater than that found in the type-I LED due to the small recombination volume localized near the interface. As Auger processes are strongly dependent on injected carrier concentration (increase as n^3), the net result is an overall increase in the Auger rate (Cn^3). We may then correctly interpret the stronger increase in the light output as a function of pressure for the type-II LED compared to the type-I LED as being associated with Auger suppression.

Finally, in type-II quantum wells, we find from theory that the radiative and Auger recombination coefficients depend on the band gap as $B \propto E_g^2$ and $C \propto E_g^{-5.5}$, respectively, and that (at zero applied pressure) $B = 10^{-10} \text{ cm}^3/\text{s}$ and $C = 8 \times 10^{-27} \text{ cm}^6/\text{s}$.⁷ This gives the radiative contribution to the current density as 14% at $P=0$ and 40% at $P=9.4 \text{ kbar}$. Simple estimates predict an increase of about 2.85 in light output when the pressure is increased from 0 to 9.4 kbar at constant current ($I=140 \text{ mA}$). This is approximately in agreement with the value of 3.74 obtained experimentally. More detailed calculations and pressure measurements on other LEDs emitting between 2–5 μm are in progress.

We are grateful to Dr N. Ahmad, Dr. S. J. Sweeney, and Dr S. Jin for assistance with the pressure measurements and useful discussions. The authors also wish to thank the EPSRC for the award of a research grant to support this work.

¹A. Krier, Philos. Trans. R. Soc. London, Ser. A **359**, 599 (2001).

²A. R. Adams, M. Silver, and J. Allam, *Semiconductors and Semimetals*, Vol. 55 (Academic, London, 1998), pp. 301–331.

³A. Krier and V. V. Sherstnev, J. Phys. D **33**, 101 (2000).

⁴H. Gao, A. Krier, and V. V. Sherstnev, *Semicond. Sci. Technol.* **14**, 441 (1999).

⁵CHLH and other valence band processes not involving the split-off band are important only in materials where Δ_{so} is very large and the valence-band mass is very light. In our case, strong mixing of the light-hole and split-off bands makes the mass heavy and reduces the corresponding Auger rate.

⁶G. G. Zegrya and A. D. Andreev, *JETP* **82**, 328 (1996).

⁷G. G. Zegrya and A. D. Andreev, *Appl. Phys. Lett.* **67**, 2681 (1995).