## Electroluminescence recombination from excited-state carrier populations in double-barrier resonant-tunneling structures

J. W. Cockburn, P. D. Buckle, M. S. Skolnick, D. M. Whittaker, W. I. E. Tagg, and R. A. Hogg Department of Physics, University of Sheffield, Sheffield S3 7RH, United Kingdom

R. Grey, G. Hill, and M. A. Pate

Science and Engineering Research Council Central Facility for III-V Materials, Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield S1 3JD, United Kingdom (Received 20 March 1992)

Electroluminescence recombination arising from electrons in both the n=1 and n=2 confined levels (E1 and E2) of the quantum well of a GaAs-Al<sub>0.4</sub>Ga<sub>0.6</sub>As p-n junction double-barrier resonant-tunneling structure is reported. At the E2 resonance, study of the relative intensities of the E2 to E1 electroluminescence permits a quantitative determination of the relative populations (1:300) of the two levels. From this the ratio of the E1 tunneling time to the intersubband scattering time is deduced. Despite the small population of E2, we show that a significant fraction of the on-resonance current still arises from tunneling through this state.

Optical spectroscopy has been shown recently to be a powerful technique for the study of transport in doublebarrier resonant-tunneling structures (DBRTS).<sup>1-5</sup> Photoluminescence (PL) intensity<sup>1</sup> and PL line-shape analysis,<sup>2</sup> magneto-optical studies,<sup>2</sup> PL excitation spectroscopy,<sup>3,4</sup> and differential absorption spectroscopy<sup>5</sup> (DAS) have all been employed to study charge build up in the quantum wells (QW's) of DBRTS. Sequential tunneling has been demonstrated from the observation of PL from electrons in the n=1 subband (E1) when the structures are biased for tunneling into the n=2 confined level (E2).<sup>2,6,7</sup>

In the present paper, electroluminescence (EL) recombination from the QW's of  $Al_{0.4}Ga_{0.6}As$ -GaAs- $Al_{0.4}Ga_{0.6}As$  DBRTS embedded in GaAs p-n junctions is reported. By comparison with PL studies of conventional n-i-n DBRTS, where the holes are supplied by photoexcitation, <sup>1-3</sup> in a p-i-n structure the holes are already present in the doped p<sup>+</sup> region. When bias is applied, both electron and hole tunneling can occur through the double-barrier region, and lead to EL recombination in the QW.<sup>8</sup>

The experiments were carried out on structures with QW's of width 70 and 80 Å which contain two confined electron levels. When biased for electron tunneling into E1, EL recombination between E1 electrons and n=1 heavy holes (HH1) ( $E_{11h}$  recombination) is observed. On increasing the bias to the second (E2) resonance, E2 electron, HH1 ( $E_{21h}$ ) recombination is also seen. This work represents a significant advance over that reported previously where only  $E_{11h}$  PL was detected. Higher sensitivity to weak signals is obtained in EL compared to PL studies; the background is zero and any optical signal detected must result from recombination in the device under study.

The structures studied were grown by molecular-beam epitaxy, and were of the following design:  $n^+$ -type GaAs substrate,  $1-\mu m$   $n=1\times 10^{18}$  cm<sup>-3</sup> GaAs, 1000-Å n=1 $\times 10^{17}$  cm<sup>-3</sup> GaAs, 50-Å undoped GaAs spacer, 80-Å Al<sub>0.4</sub>Ga<sub>0.6</sub>As barrier, 70- or 80-Å GaAs QW, 80-Å Al<sub>0.4</sub>Ga<sub>0.6</sub>As barrier, 50-Å undoped GaAs spacer, 1000-Å  $p=5\times10^{17}$  cm<sup>-3</sup> GaAs, and 9000-Å  $p=1\times10^{18}$  cm<sup>-3</sup> GaAs. The structures were processed into 100- $\mu$ m-diam mesas with annular top contacts for the collection of the EL. All the measurements were carried out with the samples immersed in liquid helium at 4.2 K.

The structures showed negligible current flow for V < 1.5 V, the flat-band condition. A forward bias of 1.5 V on the *p*-*n* junction corresponds to zero bias in a conventional n-i-n DBRTS. The I-V characteristic for the DBRTS with a 70-Å QW is shown in Fig. 1(a). Two main resonances are observed at 1.65 and 2.1 V and arise from electron tunneling into the E1 and E2 levels of the QW. These identifications are obtained by comparison with an *n-i-n* DBRTS with 80-Å barriers, and an 80-Å QW (Ref. 2), where very similar *I-V* characteristics were observed. The weak shoulders at 1.62 and 1.95 V very likely arise from hole resonant tunneling through the LH1 and LH2 levels of the QW.<sup>9,10</sup> The hole resonances are identified from the calculated confinement energies of the electron and hole levels, and from comparison with the known biases of the E1 and E2 resonances.

A series of EL spectra covering the bias ranges of the E1 and E2 resonances is shown in Fig. 2. EL is first observed at  $\sim 1.6$  V, close to the onset of the E1 and LH1 resonances. The spectrum in Fig. 2(i), taken at 1.60 V, peaks at 1.579 eV. The  $E_{11h}$  peak is calculated to occur at  $E_g(GaAs)$  (1.519 eV)+E1 (48 meV)+HH1 (13 meV)  $-E_x$  (8 meV)=1.572 eV ( $E_x$  is the exciton binding energy). This is close to the observed value of 1.579 eV.

The  $E_{11h}$  peak is observed throughout the bias range from 1.6 to 2.3 V [left-hand side, Figs. 2(i)-2(vi)]. At the onset of the E2 resonance, EL recombination is also observed at ~1.715 eV [right-hand side, Fig. 2(iv)].<sup>11</sup> The 1.715-eV emission, which is approximately 10<sup>4</sup> times weaker than that due to  $E_{11h}$ , corresponds to  $E_{21h}$  recombination. At 1.9 V the separation between the  $E_{21h}$  and  $E_{11h}$  emission peaks is 143 meV (1.716-1.573 eV), close to the calculated  $E_{2}$ -E1 separation of 195-48=147 13758



FIG. 1. (a) Current-voltage characteristics (4.2 K) showing E1 and E2 tunneling resonances at 1.65 and 2.09 V, respectively. 1.5 V corresponds to the flat-band voltage of the *p-n* junction. The weak shoulder at 1.62 V is ascribed to the LH1 resonance. (b) E1-HH1 EL intensity ( $I_1$ ) vs bias. The intensities represented by the crosses are multiplied by a factor of 40. (c) E2-HH1 EL intensity ( $I_2$ ) vs bias. (d) Ratio of  $I_2/I_1$  EL intensities vs bias. The vertical dashed line at 1.9 V signifies the onset of the region above 1.9 V where the  $I_2/I_1$  ratio is not affected by the nonzero E1 population below the E2 resonance.

meV, confirming the identification of the 1.715-eV peak. At the E2 resonance, electrons tunnel into the n=2 electron state. As shown in Fig. 3, they can then either tunnel directly out of the well (rate  $1/\tau_2$ ) or they can undergo intersubband scattering (rate  $1/\tau_i$ ) by rapid LO phonon emission to E1, accumulate in E1, and then tunnel out of the well (rate  $1/\tau_1$ ).

The  $E_{11h}$  and  $E_{21h}$  integrated intensities ( $I_1$  and  $I_2$ , respectively) are plotted as a function of bias in Figs. 1(b) and 1(c). Between 1.6 and 1.85 V only  $E_{11h}$  is observed. A sharp peak in  $I_1$  is observed at 1.62 V, corresponding to the shoulder seen in *I*-V arising from tunneling into LH1. The EL intensity is controlled by both the electron and hole populations in the QW, <sup>1,2,8</sup> and so resonances due to both electron and hole tunneling are expected.<sup>8</sup> The results in Figs. 1(a) and 1(b) show that the EL intensity is a much more sensitive function of the hole population in the QW than the tunnel current, in agreement with previous PL results. <sup>12,13</sup> No feature corresponding to the *E*1 resonance in *I*-V [Fig. 1(a)] is seen in Fig. 1(b). This is not well understood, but in the 80-Å well sample  $I_1$  does peak



FIG. 2. EL spectra at 4.2 K as a function of bias. E1-HH1 is presented in the left half of the figure and E2-HH1 in the right half. Between 1.60 and 1.64 V [(i)-(iii)] only E1-HH1 EL is observed. Beyond the onset of the E2 resonance at 1.86 V both E1-HH1 and E2-HH1 signals are seen [(iv)-(vi)].



FIG. 3. Schematic diagram of the DBRTS at 2.0 V applied bias. The rates for tunneling out from E2 and E1  $(1/\tau_2 \text{ and } 1/\tau_1)$  and the intersubband scattering rate  $1/\tau_i$  are indicated.

13759

at the E l resonance. It is most likely that the E l resonance in  $I_1$  in the 70-Å QW is suppressed by the rapidly decreasing hole population in this bias range.

We now direct attention to the behavior of the EL intensities  $(I_1, I_2)$  in Figs. 1(b) and 1(c) beyond 1.85 V. A good correlation between  $I_1(V)$  and  $I_2(V)$  is seen in Figs. 1(b) and 1(c). Both show a very strong increase at 1.85 V, the onset of the E2 resonance in I-V in Fig. 1(a). For V < 1.85 V,  $I_2$  is zero. By contrast  $I_1$  is nonzero in the 1.68-1.85 V region due to the occurrence of inelastic tunneling [the broad peak at 1.72 V in Fig. 1(a) is due to LO phonon-assisted tunneling]. Both  $I_1$  and  $I_2$  peak at ~1.97 V before decreasing towards the cutoff of the resonance at  $\sim 2.1$  V. The peak in  $I_2$  and  $I_1$  occurs at 1.97 V, even though the electron populations must increase up to the cutoff of the resonance at 2.1 V, because of the decreasing HH1 population beyond 1.97 V. An analogous result was found in PL on the very similar *n-i-n* structure of Ref. 2 beyond 0.4 V. As explained in Ref. 2, beyond 0.4 V (1.9 V for a *p-i-n* structure) holes which tunnel through the first barrier have sufficient energy to pass directly over the top of the second barrier without capture by the QW.

The ratio  $I_2/I_1$ , in the region of the second resonance, is plotted in Fig. 1(d). The values below 1.9 V are affected by the fact that  $I_1$  is nonzero at lower biases where  $E_2$ tunneling cannot occur and  $I_2=0$  [the region to the left of the dashed vertical line of Fig. 1(d)]. In the main region of interest from 1.9 to 2.2 V the  $I_2/I_1$  ratio varies by less than a factor of 1.7 with a minimum value of  $1.2 \times 10^{-4}$  at 2.0 V. A very similar value is found for the 80-Å QW DBRTS. Since both  $E_2$  and  $E_1$  electrons recombine with HH1 holes, the ratio  $I_2/I_1$  is independent of the hole population, and is given by

$$\frac{I_2}{I_1} = \frac{n_2}{n_1} \frac{f_2}{f_1} \,, \tag{1}$$

where  $n_2$  and  $n_1$  are the E2 and E1 populations, respectively, and  $f_2/f_1$  is the ratio of oscillator strengths for  $E_{21h}$ to  $E_{11h}$  transitions.<sup>14</sup> The  $f_2/f_1$  ratio was calculated from a solution of Schrödinger's equation for the QW, using electric-field values obtained from solution of Poisson's equation. At 2.0-V bias  $f_2/f_1$  is calculated to be 0.08, increasing to 0.1 at 2.1 V. Some absorption of the EL will occur in the top GaAs contacts. This is a small effect, but will lead to attenuation of  $I_2$  relative to  $I_1$  by a factor of about 2.<sup>15</sup> Using the 2.0-V values of  $I_2/I_1 = 1.2 \times 10^{-4}$  $(2.4 \times 10^{-4}$  after accounting for GaAs attenuation) and  $f_2/f_1 = 0.08$ ,  $n_2/n_1$  is obtained from Eq. (1) to be  $3 \times 10^{-3}$ .

When the structure is biased at the E2 resonance,  $n_1$ and  $n_2$  are determined by the following rate equations:

$$\frac{dn_2}{dt} = G - \frac{n_2}{\tau_2} - \frac{n_2}{\tau_i} - \frac{n_2}{\tau_{R2}}, \qquad (2)$$

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_i} - \frac{n_1}{\tau_1} - \frac{n_1}{\tau_{R1}},$$
(3)

where G is a "generation" rate for  $n_2$  electrons determined by tunneling in from the emitter contact  $\tau_2$ ,  $\tau_1$ , and  $\tau_i$ were defined earlier (see Fig. 3), and  $(\tau_{R2})^{-1}$  and  $(\tau_{R1})^{-1}$  are radiative recombination rates. In the steady state, from Eq. (3),

$$\frac{n_2}{\tau_i} - \frac{n_1}{\tau_1} - \frac{n_1}{\tau_{R1}} = 0, \qquad (4)$$

and so  $n_2/n_1 = \tau_i(1/\tau_1 + 1/\tau_{R1})$ . Electrons relax from E2 to E1 by rapid LO phonon intersubband scattering at a rate  $1/\tau_i \sim 2 \times 10^{12}$  sec<sup>-1</sup>.<sup>16</sup> Using the deduced value of  $n_2/n_1$  of  $3 \times 10^{-3}$ ,  $1/\tau_1 + 1/\tau_{R1}$  is thus found to be  $6 \times 10^9$  $sec^{-1}$ . At the probable carrier densities in the well  $(< 10^{10} \text{ cm}^{-2} \text{ for at least one of the carriers away from}$ the peak of resonance)  $\tau_{R1}^{-1}$  will very likely be < 10<sup>8</sup> sec<sup>-1</sup>, and can be neglected relative to  $\tau_1^{-1.17}$  As a result  $\tau_1^{-1}$  is found to be  $6 \times 10^9$  sec<sup>-1</sup>. Calculated values for  $\tau_1^{-1}$  are extremely sensitive to the barrier width and height employed. For example, for d = 80 Å, we obtain a value for  $\tau_1^{-1}$  of  $1.1 \times 10^9$  sec<sup>-1</sup>, from the calculated width of the  $E_1$  resonance at 2.0 V applied bias, increasing to  $4.6 \times 10^9$  sec<sup>-1</sup> for d = 70 Å. Bearing in mind this sensitivity to the input parameters we conclude that the experimental value of  $\tau_1^{-1}$  of  $6 \times 10^9$  sec<sup>-1</sup> is in very reasonable agreement with the calculated values, and provides confidence in the analysis of  $I_2/I_1$  intensities.

The rate equations show that the  $n_2/n_1$  population ratio is determined by  $\tau_i/\tau_1$ .  $n_2/n_1$  is small ( $\sim 3 \times 10^{-3}$ ) due to the rapid intersubband scattering by LO phonons ( $\hbar\omega \sim 36$  meV) between states  $\sim 150$  meV apart in energy. By contrast the absolute values of  $n_2$  and  $n_1$  are also determined by the tunneling in (G) and tunneling out  $(1/\tau_2)$  rates from E2 [see Eq. (2)]. We calculate  $1/\tau_2$  to be  $1 \times 10^{12}$  sec<sup>-1</sup>, for d = 80 Å at 2-V bias. The ratio of the E2 to E1 current densities is given by  $J_2/J_1$  $= (n_2/n_1)(\tau_1/\tau_2)$ , which is  $\sim 3$  for  $n_2/n_1 = 3 \times 10^{-3}$ ,  $\tau_1/\tau_2 \approx 10^3$ . Thus, even though the  $n_2/n_1$  ratio is very small it is clear that a significant part of the tunnel current is carried by tunneling through the E2 state, with the rest being transported by a sequential process via E 1.<sup>18</sup>

The above analysis of the  $I_2/I_1$  and  $n_2/n_1$  ratios has been performed at 2.0-V applied bias. The reason for the rapidly increasing  $I_2/I_1$  ratio for V = 1.86 to 1.90 V has been discussed earlier. The decrease of  $I_2/I_1$  from 1.9 to 2.0 V is not well understood. However, beyond 2.0 V, where  $I_2/I_1$  increases by a factor of 1.7 from 2.0 to 2.1 V, the increase can be understood in terms of the decreasing value of  $\tau_1$  with bias [see Eq. (1) and (4)], and of the expected increase of  $f_2/f_1$  with bias. We calculate a decrease of  $\tau_1$  by a factor of 2, from 2 to 2.1 V. This is a larger variation than that expected for the increase of the  $f_2/f_1$  ratio (of a factor of 1.3) over the same bias range, and can account at least qualitatively for the increase in  $I_2/I_1$  observed in Fig. 1(d). When the device goes off resonance at 2.1 V,  $I_2/I_1$  decreases again. This is expected since the electric field across the collector barrier will decrease, and hence  $\tau_1$  will increase as charge is ejected from the well between the off- and on-resonance states.

Related results have been reported by Grahn *et al.* in PL experiments on GaAs-AlAs superlattices.<sup>14</sup> In those experiments tunneling occurred by a sequential process from E1 to E2 of adjacent wells, followed by intersubband scattering from E2 to E1 and then tunneling to E2 in the next well.<sup>19</sup> The present case is much more straightforward to analyze since tunneling through only 13760

one QW is involved.

To conclude, EL recombination from the ground and first excited electron state of the QW of a p-n junction DBRTS has been reported. Study of the relative intensities of E2 to E1 recombination at the E2 resonance has permitted the ratio of the E1 tunneling time to the intersubband scattering time to be deduced. In spite of the small E2 population deduced, a significant fraction of the

- <sup>1</sup>J. F. Young, B. M. Wood, G. C. Aers, R. L. S. Devine, H. C. Liu, D. Landheer, M. Buchanan, A. J. Springthorpe, and P. Mandeville, Phys. Rev. Lett. **60**, 2085 (1988).
- <sup>2</sup>M. S. Skolnick, D. G. Hayes, P. E. Simmonds, A. W. Higgs, G. W. Smith, H. J. Hutchinson, C. R. Whitehouse, L. Eaves, M. Henini, O. H. Hughes, M. L. Leadbeater, and D. P. Halliday, Phys. Rev. B 41, 10754 (1990).
- <sup>3</sup>M. S. Skolnick, P. E. Simmonds, D. G. Hayes, A. W. Higgs, G. W. Smith, A. D. Pitt, C. R. Whitehouse, H. J. Hutchinson, C. R. H. White, L. Eaves, M. Henini, and O. H. Hughes, Phys. Rev. B 42, 3069 (1990).
- <sup>4</sup>H. Yoshimura, J. N. Schulman, and H. Sakaki, Phys. Rev. Lett. **64**, 2422 (1990).
- <sup>5</sup>T. K. Woodward, D. S. Chemla, I. Bar-Joseph, H. U. Baranger, D. L. Sivco, and A. Y. Cho, Phys. Rev. B 44, 1353 (1991).
- <sup>6</sup>A similar conclusion has been reached by Woodward *et al.* in Ref. 5 using differential absorption spectroscopy.
- <sup>7</sup>Sequential tunneling for tunneling into E1 has been demonstrated for a structure with a thick collector barrier by M. L. Leadbeater, E. S. Alves, F. W. Sheard, L. Eaves, M. Henini, O. H. Hughes, and G. A. Toombs, J. Phys. Condens. Matter 1, 10605 (1989). The PL results of Ref. 2 support this conclusion.
- <sup>8</sup>C. van Hoof, J. Genoe, R. Merlens, G. Borghs, and E. Goovaerts, Appl. Phys. Lett. **60**, 77 (1991).
- <sup>9</sup>E. E. Mendez, L. Esaki, and W. I. Wang, Phys. Rev. B 33, 2893 (1986).

on-resonance current is carried by tunneling through E2, with the rest being transported by sequential processes via E1.

Financial support for this work from the Science and Engineering Research Council, United Kingdom, is gratefully acknowledged. We wish to thank L. Eaves for a very helpful discussion.

- <sup>10</sup>As discussed in R. K. Hayden, D. K. Maude, L. Eaves, E. C. Valadares, M. Henini, F. W. Sheard, O. H. Hughes, J. C. Portal, and L. Cury, Phys. Rev. Lett. **66**, 1749 (1991), the tunneling probability through HH1 is too small to lead to an observable peak in *I-V*.
- <sup>11</sup>Weak emission at 1.599 eV is observed arising from an *E*1-LH1 recombination, and will be discussed elsewhere.
- <sup>12</sup>N. Vodjdani, D. Cote, D. Thomas, B. Sermage, P. Bois, E. Costard, and J. Nagle, Appl. Phys. Lett. 56, 33 (1990).
- <sup>13</sup>C. R. H. White, M. S. Skolnick, P. E. Simmonds, L. Eaves, M. Henini, O. H. Hughes, G. Hill, and M. A. Pate, Superlattices Microstruct. 8, 195 (1990).
- <sup>14</sup>H. T. Grahn, H. Schneider, W. W. Ruhle, K. von Klitzing, and K. Ploog, Phys. Rev. Lett. 64, 2426 (1990).
- <sup>15</sup>M. D. Sturge, Phys. Rev. **127**, 768 (1962).
- <sup>16</sup>M. C. Tatham, J. F. Ryan, and C. T. Foxon, Phys. Rev. Lett. **63**, 1637 (1989). An upper limit for  $\tau_i$  of  $10^{-12}$  sec was measured in this work.
- <sup>17</sup>T. Matsusue and H. Sakaki, Appl. Phys. Lett. **50**, 1429 (1987).
- <sup>18</sup>M. L. Leadbeater, L. Eaves, M. Henini, O. H. Hughes, G. Hill, and M. A. Pate, Solid-State Electron. **32**, 1467 (1989).
- <sup>19</sup>Population of the excited states of a superlattice has been demonstrated by M. Helm, P. England, E. Colas, F. DeRosa, and S. J. Allen, Phys. Rev. Lett. 63, 74 (1989), from the detection of infrared emission resulting from intersubband relaxation.