PHYSICAL REVIEW B VOLUME 57, NUMBER 11 15 MARCH 1998-I

Photoluminescence spectroscopy of intersubband population inversion in a GaAs/Al$_x$Ga$_{1-x}$As triple-barrier tunneling structure

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(Received 23 June 1997)

We have used interband photoluminescence (PL) spectroscopy to demonstrate the occurrence of an intersubband population inversion in a GaAs/Al$_x$Ga$_{1-x}$As triple barrier structure containing asymmetric coupled quantum wells. The relative populations of the $n=1$ ($E1$) and $n=2$ ($E2$) electron subbands of the wider quantum well (QW1) are deduced from the relative intensities of the PL peaks arising from recombination of the $E1$ and $E2$ electrons. A significant population inversion is obtained between $E2$ and $E1$ when the structure is biased so that $E1$ is in resonance with the $n=1$ ($E1*$) level of the narrower quantum well (QW2). The key importance of the interwell resonance in achieving population inversion is confirmed by comparison with PL results from structures in which $E1$-$E1*$ alignment does not occur. [S0163-1829(98)03608-X]

There is currently considerable interest in the study of midinfrared (IR) sources based on electronic intersubband transitions within the quantum wells (QWs) of semiconductor low dimensional structures.[1–3] Continued development of such devices requires a soundly based understanding of the effects of changing design parameters on the intersubband population kinetics. Techniques such as differential absorption spectroscopy and photocurrent spectroscopy have provided useful information on the bias and doping dependence of intersubband transition energies and line shapes in quantum cascade (QC) intersubband emitters.[4,5] However, these experiments have not addressed one of the most fundamental issues, namely the dependence of the relative subband populations on device design and operating conditions.

In previous work[6–9] we have shown that *interband* luminescence spectroscopy may be used as a direct, quantitative probe of electron subband populations. We have demonstrated the dependence of ground and excited state populations on tunneling rates and intersubband scattering rates in double barrier structures, and shown that intersubband population inversions can be obtained in appropriately designed devices.[8,9] In the present paper we extend the technique to the study of subband populations in triple barrier resonant tunneling structures (TBRTS) incorporating asymmetric coupled QWs. Such structures are very similar in concept to the active regions of the vertical transition QC laser.[2]

A schematic band-edge diagram of a TBRTS, biased for tunneling into the $n=2$ ($E2$) level of the wide QW (QW1), is shown in Fig. 1(a). At such biases, the relative populations of the $E2$ and $E1$ subbands are given by $n_2/n_1 \approx \tau_{21}/\tau_1$, where τ_{21} is the LO-phonon mediated $E2$-$E1$ intersubband scattering time and τ_1 is the tunneling out time from $E1$.[6] In order to achieve population inversion between $E2$ and $E1$, the QW widths are chosen so that the $E1$ level of QW1 is energetically aligned with the ground state ($E1*$) of the narrow well (QW2) at the $E2$ resonance bias.[10] $E1$ electrons may thus escape rapidly by resonant tunneling via $E1*$, on a time scale shorter than the intersubband scattering time (~ 1 ps).[11] Since $E2$ electrons must tunnel nonresonantly through a relatively wide region comprising the intermediate and collector barriers and QW2, the $E2$ escape time is significantly longer. Such structures therefore provide the short $E1$ lifetime necessary to achieve $E2$-$E1$ population inversion, while maintaining a high density of $E2$ electrons available for intersubband transitions.

In the present paper we concentrate on the results of interband photoluminescence (PL) experiments on a TBRTS with QW1 and QW2 widths of 66 and 33 Å, respectively (66/33 structure). Poisson-Schrödinger calculations for this structure predict the $E1$ and $E1*$ levels to be energetically aligned when the device is biased for tunneling into the $E2$ QW1 state. A strong enhancement of the n_2/n_1 population ratio is therefore expected to result. Analysis of the relative intensities of the PL peaks arising from recombination of $E2$ and $E1$ electrons with heavy holes from the $n=1$ (HH1) QW1 state confirms this prediction, indicating a significant intersubband population inversion at the $E1$-$E1*$ resonance bias. Comparative experiments on structures in which the QW ground states are not expected to be in resonance show that $n_1 > n_2$ at all biases, confirming the key role played by the interwell resonance in achieving population inversion. The results clearly demonstrate the ability of interband PL spectroscopy to provide detailed, quantitative information on population kinetics in intersubband devices.

The 66/33 structure, which was grown by molecular beam epitaxy on a semi-insulating GaAs substrate, consisted of the following: 2 μm $n=7\times10^{17}$ cm^{-3} GaAs buffer layer, 1000 Å $n=1\times10^{17}$ cm^{-3} GaAs emitter, 100 Å undoped GaAs spacer, 66 Å undoped Al$_{0.4}$Ga$_{0.6}$As emitter barrier, 66 Å undoped GaAs QW1, 26 Å undoped Al$_{0.4}$Ga$_{0.6}$As intermediate barrier, 33 Å undoped GaAs QW2, 26 Å undoped Al$_{0.4}$Ga$_{0.6}$As collector barrier, 100 Å undoped GaAs spacer, 1000 Å $n=1\times10^{17}$ cm^{-3} GaAs collector, 0.4 μm $n=7\times10^{17}$ cm^{-3} GaAs top contact. The widths of QW1 and

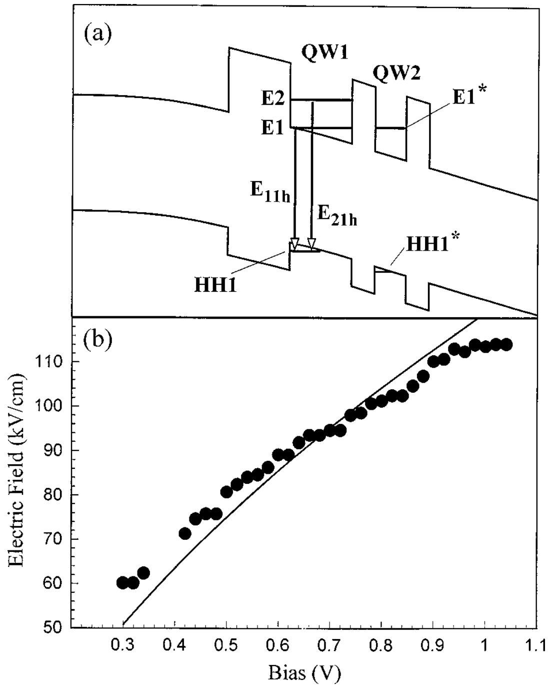

FIG. 1. (a) Schematic band-edge diagram of TBRTS biased for tunneling into the $E2$ level of the wide quantum well (QW1). (b) Calibration of electric field against device bias obtained from magnetotransport measurements of the sheet density in the electron accumulation layer. The solid line shows the predicted calibration, based on the solution of Poisson-Schrödinger equations for the structure.

QW2 were confirmed to be 66 and 33 Å, respectively, from measurements of the $E1$-HH1 and $E1^*$-HH1* transition energies by PL excitation spectroscopy.

Under forward bias, photocreated holes from the top contact are driven to the triple barrier region. Since the hole interwell tunneling time is expected to be short (~ 10 ps for a 30 Å intermediate barrier[12,13]) the hole population in QW2 will be negligible, with most of the holes relaxing to the HH1 level of QW1 before tunneling out of the structure. These holes recombine with $E1$ and $E2$ electrons to generate the PL features of interest. Following the procedure adopted in Ref. 14, we estimate an upper limit of 10^7 cm^{-2} for the HH1 hole density, about three orders of magnitude less than the maximum expected electron density in QW1.[15] At these relatively low carrier densities, the PL intensity is proportional to both the electron and hole concentrations within the relevant subbands, irrespective of whether the recombination is free particle or excitonic in nature.[14] Since both $E1$ and $E2$ electrons recombine with the same HH1 holes, the ratio of the $E2$-HH1 and $E1$-HH1 PL intensities, from which the n_2/n_1 population ratio is deduced, is unaffected by variations in the HH1 population.

The I-V characteristic obtained from the structure at a temperature of 2 K is shown in Fig. 2(a). Two main resonances are observed, corresponding to electrons from the emitter accumulation layer tunneling into the $E1$ and $E2$ levels of QW1. In the bias range between the peaks of the $E1$ and $E2$ resonances, an additional peak [labeled $E1^*$ in Fig. 2(a)] is observed. We attribute this feature to resonant tunneling of emitter electrons through the $E1^*$ confined level of QW2.[16] To facilitate data analysis, the electric field within the structure was calibrated against device bias by means of Shubnikov–de Haas–like magnetotransport measurements of the electron sheet density in the emitter accumulation layer[17] [Fig. 1(b)].

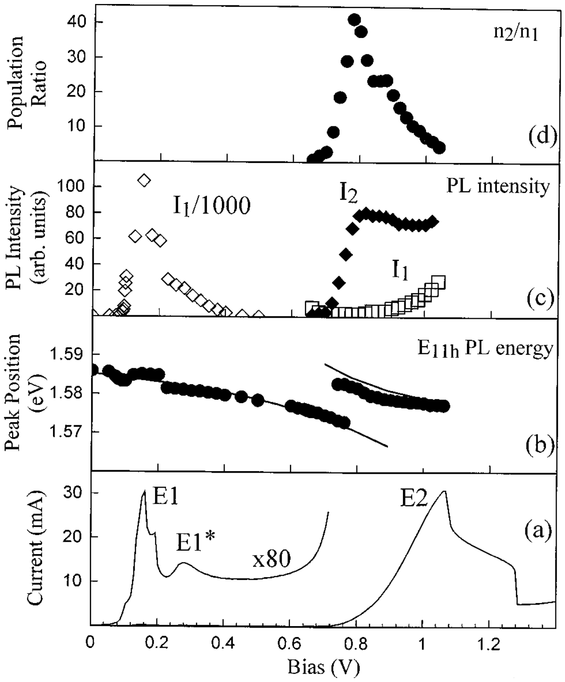

FIG. 2. (a) I-V characteristic, (b) E_{11h} PL energy versus bias, (c) E_{11h} and E_{21h} PL intensities versus bias, (d) n_2/n_1 population ratio versus bias, obtained from TBRTS with QW1 = 66 Å and QW2 = 33 Å.

The PL spectra (2 K) obtained from the structure as a function of forward bias are shown in Fig. 3(a). The peak labeled E_{11h}, which emerges at the onset of the $E1$ resonance in I-V, is due to the recombination of $E1$ electrons with HH1 holes. When the bias is increased beyond the onset of the $E2$ resonance, an additional PL peak emerges due to $E2$-HH1 (E_{21h}) recombination.[6] At the bias corresponding to the peak of the $E2$ resonance (1.05 V), the energy separation between the E_{11h} and E_{21h} PL is 154 meV, very close to the calculated $E2$-$E1$ splitting of 156 meV for a 66 Å QW under an applied electric field of 120 kV cm^{-1}.

The bias dependence of the E_{11h} spectra is particularly interesting in the 0.7–0.9 V range. Spectra obtained from around this region are plotted on an expanded scale in Fig. 4. At 0.62 V a single E_{11h} peak is observed at 1.577 eV. As the bias is increased, the peak shifts to lower energy due to the quantum confined Stark effect. The peak also decreases significantly in intensity, and at biases of around 0.8 V splits into two very weak components (E_{11h}^- and E_{11h}^+) separated in energy by approximately 10 meV. This quenching of the PL intensity is consistent with the significant reduction of the $E1$ escape time, which is expected at the $E1$-$E1^*$ resonance. As the bias is further increased towards the peak of the $E2$ resonance, the higher energy feature increases significantly in intensity and Stark shifts to lower energy.

This splitting of E_{11h} into two components is fully consistent with the splitting which is expected to occur between the symmetric and antisymmetric states formed when the $E1$ and $E1^*$ ground states of QW1 and QW2 are resonantly coupled.[18,19] For the present structure, the symmetric-

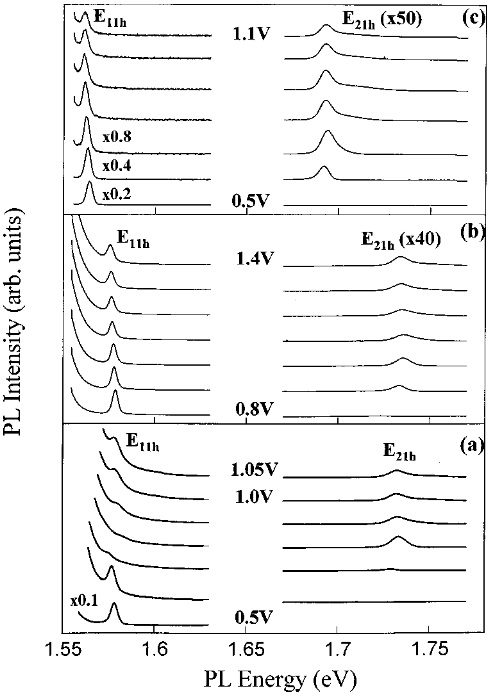

FIG. 3. E_{11h} and E_{21h} PL spectra obtained in the bias range of the $E2$ resonance from (a) the 66/33 structure, (b) the 66/20 structure and (c) the 80/30 structure. The bias increases in increments of 0.1 V between spectra, except where indicated. Note the multiplication factors in the E_{21h} intensities in (b) and (c).

antisymmetric splitting is calculated to be 13 meV, in good agreement with the observed peak splitting of approximately 10 meV. Beyond the $E1$-$E1^*$ resonance, the $E1$ and $E1^*$ states again become increasingly localized in QW1 and QW2, respectively. Only E_{11h} PL is then observed in this energy range, due to the relatively low oscillator strength of the $E1^*$-HH1 transition. The bias dependence of the E_{11h} PL peak position is plotted in Fig. 2(b). Also shown (solid line) is the calculated E_{11h} peak position obtained from envelope function calculations, assuming an E_{11h} exciton binding energy of 9 meV. The calculated bias dependence agrees closely with the experimental data throughout the bias range of the experiment. In particular, it shows that an anticrossing of the $E1$ and $E1^*$ levels is expected at biases of around 0.8 V, providing strong support for the above interpretation.

The integrated intensities of the E_{11h} and E_{21h} PL peaks (I_1 and I_2, respectively) are plotted versus bias in Fig. 2(c).[20] Throughout the bias range of the $E2$ resonance, I_2 is significantly greater than I_1. The ratio of the $E2$ and $E1$ populations probed by the PL measurements is obtained from the PL intensity ratio, after correcting for the energy dependence of PL reabsorption in the GaAs top contact, the spectral response of the measurement system, and the relative oscillator strengths of the E_{21h} and E_{11h} transitions.[6–9] The deduced n_2/n_1 ratio is plotted versus vias in Fig. 2(d). The data are subject to an error of around $\pm 30\%$, based on estimated uncertainties in growth parameters, PL intensity measurements, and oscillator strength calculations. The results show that $n_2 > n_1$ over most of the $E2$ resonance, with a sharp maximum occurring at the $E1$-$E1^*$ resonance bias of around 0.8 V, where $n_2/n_1 \approx 40$.

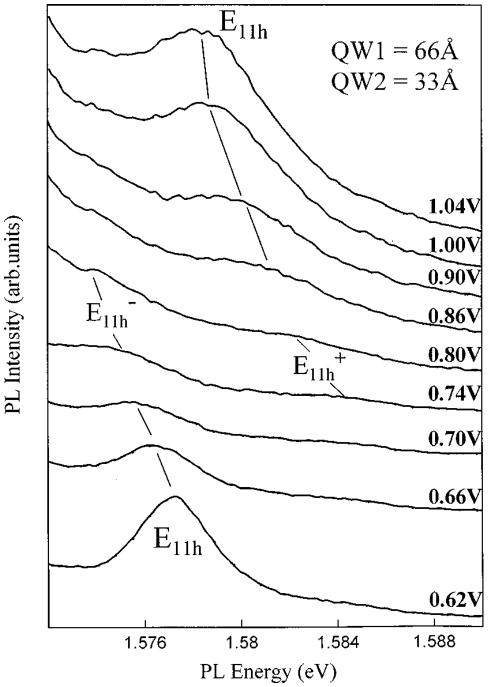

FIG. 4. PL spectra in the E_{11h} energy range for biases around the $E1$-$E1^*$ resonance, showing the symmetric-antisymmetric splitting, which occurs at around 0.8 V.

Such a large population ratio is clearly inconsistent with the relation $n_2/n_1 \approx \tau_{21}/\tau_1$, since it would require an unrealistically short $E1$ lifetime. Following the method of Ref. 11 to calculate the scattering time from $E2$ to the coupled $E1$-$E1^*$ states, we find $\tau_{21} \approx 1$ ps. Due to the delocalization of the wave function at the $E1$-$E1^*$ resonance, the escape time from the coupled $E1$-$E1^*$ states is twice as long as the tunneling out time from the uncoupled $E1^*$ state.[21] We calculate the $E1^*$ tunneling out time to be 0.15 ps,[22] giving an escape time of approximately 0.3 ps from the coupled $E1$-$E1^*$ states. These values for τ_{21} and τ_1 give a predicted population ratio of $n_2/n_1 \approx 3$ at the $E1$-$E1^*$ resonance.

In order to understand this large discrepancy between the measured and predicted population ratios ($n_2/n_1 \approx 40$ and 3, respectively), it must be remembered that it is the *overall* n_2/n_1 population ratio which is given by the ratio of τ_{21} to τ_1. The PL measurements, however, sample only those electronic states that contribute to the formation of the optically active exciton wave function. This involves electrons which occupy states within about one exciton binding energy (~ 9 meV) of the subband minimum. Since the $E1$-$E1^*$ resonance occurs close to the onset of the $E2$ resonance in I-V, electrons are injected into the $E2$ subband with low in-plane energy, and we therefore expect the entire $E2$ population to be probed by the PL measurements.

By contrast, electrons which relax from the bottom of the $E2$ subband by LO phonon emission enter the $E1$ subband with excess in-plane energy of $E2$-$E1$-$\hbar\omega_{\mathrm{LO}} \approx 118$ meV. Using a model similar to that proposed by Faist *et al.*,[2] we ignore carrier-carrier interactions (which are negligible at the low carrier densities present in our structure) and assume that these electrons relax within $E1$ by sequential emission of LO phonons. At any point in the relaxation cascade, $E1$ electrons may either tunnel out of $E1$ at a rate $1/\tau_1$ or undergo intrasubband relaxation with the emission of LO phonons at a rate $1/\tau_i$. It can then be shown from steady-state rate equation analysis that the fraction of the total $E1$ population which relaxes to the subband minimum, and is therefore accessible to the PL measurements, is given by

$$n_1^{\mathrm{PL}} = n_1 \left(\frac{\tau_1}{\tau_1 + \tau_i} \right)^N, \tag{1}$$

where N is the number of sequential LO phonon emission steps between the $E1$ injection energy and the subband minimum. Ferreira and Bastard[11] calculate the intrasubband LO-phonon emission time τ_i to be ~ 300 fs, which is approximately equal to our calculated $E1$ lifetime. For an $E1$ injection energy of 118 meV we have $N=3$, and we thus estimate $n_1^{\mathrm{PL}} = n_1/8$ from Eq. (1). The maximum value of $n_2/n_1^{\mathrm{PL}} \approx 40$ obtained from the PL measurements therefore corresponds to an *overall* population ratio of $n_2/n_1 \approx 5$, in very reasonable agreement with the predicted estimate of $n_2/n_1 \approx 3$.

To further confirm the importance of the $E1$-$E1^*$ resonance in producing the population inversion, two additional structures with narrower QW2 widths were studied. The first of these had a QW1 of 66 Å width, and a 20 Å wide QW2 (66/20 structure), while the second had well widths of 80 and 30 Å, respectively (80/30 structure). In both structures, the $E1^*$ level is expected to remain higher in energy than $E1$ throughout the bias range of the experiment. PL spectra obtained from the 66/20 and 80/30 samples at biases in the range of the $E2$ resonance are plotted in Figs. 3(b) and 3(c), respectively. For both structures, I_2 is significantly less than I_1 at all biases, with the maximum intensity ratio being obtained at the peak of the $E2$ resonance, where $I_2/I_1 \approx 0.1$. From Eq. (1) we obtain a maximum population ratio of $n_2/n_1 \approx 0.3$ for both structures, very much less than the ratio found for the 66/33 structure in which the $E1$-$E1^*$ resonance occurs.

In summary, we have carried out a PL investigation of intersubband population kinetics in GaAs/Al$_x$Ga$_{1-x}$As TBRTS. When a structure is biased so that the ground states of the wide and narrow QWs are in resonance, a population inversion is obtained between the $E1$ and $E2$ subbands of the wide QW. The results provide an unambiguous illustration of the sensitivity of subband population ratios to relatively small changes in device design, and clearly demonstrate the power of interband PL spectroscopy as a quantitative probe of electron population dynamics in intersubband devices.

We are grateful to D. M. Whittaker for assistance with the calculation of intersubband scattering times.

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[1] J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science **264**, 553 (1994).

[2] J. Faist, F. Capasso, C. Sirtori, D. L. Sivco, A. L. Hutchinson, M. S. Hybertsen, and A. Y. Cho, Phys. Rev. Lett. **76**, 411 (1996).

[3] S. Sauvage, Z. Moussa, P. Boucaud, F. H. Julien, V. Berger, and J. Nagle, Appl. Phys. Lett. **70**, 1345 (1997).

[4] J. Faist, F. Capasso, C. Sirtori, D. Sivco, A. L. Hutchinson, S. N. G. Chu, and A. Y. Cho, Appl. Phys. Lett. **64**, 1144 (1994).

[5] J. Faist, F. Capasso, C. Sirtori, D. Sivco, A. L. Hutchinson, S. N. G. Chu, and A. Y. Cho, Appl. Phys. Lett. **65**, 94 (1994).

[6] J. W. Cockburn, P. D. Buckle, M. S. Skolnick, D. M. Whittaker, W. I. E. Tagg, R. A. Hogg, R. Grey, G. Hill, and M. A. Pate, Phys. Rev. B **45**, 13 757 (1992).

[7] P. D. Buckle, J. W. Cockburn, R. J. Teissier, A. R. K. Willcox, D. M. Whittaker, M. S. Skolnick, G. W. Smith, R. Grey, G. Hill, and M. A. Pate, Semicond. Sci. Technol. **9**, 533 (1994).

[8] J. W. Cockburn, M. S. Skolnick, D. M. Whittaker, P. D. Buckle, A. R. K. Willcox, and G. W. Smith, Appl. Phys. Lett. **64**, 2400 (1994).

[9] M. J. Birkett, J. W. Cockburn, M. S. Skolnick, D. M. Whittaker, R. Grey, G. Hill, M. A. Pate, and G. W. Smith, in *Proceedings of the 23rd International Conference on the Physics of Semiconductors*, edited by M. Scheffler and R. Zimmerman (World Scientific, Singapore, 1996), p. 2267.

[10] A. Kastalsky, V. J. Goldman, and J. H. Abeles, Appl. Phys. Lett. **59**, 2636 (1991).

[11] R. Ferreira and G. Bastard, Phys. Rev. B **40**, 1074 (1989).

[12] B. Deveaud, A. Chomette, F. Clerot, P. Auvray, A. Regreny, R. Ferreira, and G. Bastard, Phys. Rev. B **42**, 7021 (1990).

[13] M. Nido, M. G. W. Alexander, W. W. Rühle, T. Schweizer, and K. Köhler, Appl. Phys. Lett. **56**, 355 (1990).

[14] 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).

[15] 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**, 10 754 (1990). In this paper PL line-shape analysis and magneto-PL experiments are used to deduce values for the electron densities in double barrier structures having longer carrier lifetimes than the present TBRTS. These values are used to estimate upper limits for the electron densities in the present experiment.

[16] This attribution is confirmed by comparison with the I-V characteristics obtained from other triple barrier structures, in which the peak bias of the $E1^*$ resonance is observed to increase systematically as the width of QW2 is decreased.

[17] L. Eaves, G. A. Toombs, F. W. Sheard, C. A. Payling, M. L. Leadbeater, E. S. Alves, T. J. Foster, P. E. Simmonds, M. Henini, O. S. Hughes, J. C. Portal, G. Hill, and M. A. Pate, Appl. Phys. Lett. **52**, 212 (1988).

[18] J. E. Golub, P. F. Liao, D. J. Eilenberger, J. B. Harbison, L. T. Florez, and Y. Prior, Appl. Phys. Lett. **53**, 2584 (1988).

[19] A. M. Fox, D. A. B. Miller, G. Livescu, J. E. Cunningham, J. E. Henry, and W. Y. Jan, Phys. Rev. B **42**, 1841 (1990).

[20] The E_{21h} PL intensity saturates at a bias of around 0.8 V even though the $E2$ population continues to increase up to the peak of the $E2$ resonance. This occurs because the hole capture rate in the QW decreases significantly when the potential drop across the collector barrier and QW region exceeds the height of the valence-band emitter barrier (Refs. 14 and 15). Since both $E2$ and $E1$ electrons recombine with the same HH1 population, the intensity ratio is unaffected by this behavior.

[21] S. A. Gurvitz, I. Bar-Joseph, and B. Deveaud, Phys. Rev. B **43**, 14 703 (1991).

[22] I. A. Larkin and Y. N. Khanin, Semiconductors **27**, 990 (1993).