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Tunnelling dynamics of holes in GaAs/$Al_{0.33}Ga_{0.67}As$ double-barrier resonant tunnelling structures studied by time-resolved photoluminescence spectroscopy

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Abstract. We have studied a biased double-barrier resonant tunnelling structure using time-resolved and continuous-wave photoluminescence (PL) spectroscopy and identified the principal mechanisms which contribute to the evolution of luminescence from the quantum well (QW) region. We find that in the structures investigated the PL intensity is dominated by the diffusion and tunnelling of minority holes, in contrast with several earlier studies in which the electron behaviour was suggested to control the PL characteristics. The processes dominating the variation of PL intensity with bias are: field-driven accumulation of holes in a layer adjacent to one barrier, tunnelling of holes from this layer into the QW and the escape of holes from the QW by tunnelling at low bias, and at high bias by direct escape over the emitter barrier. Additionally, the absence of luminescence corresponding to recombination from the upper electron level when the device is biased at the second tunnelling resonance implies fast inter-subband scattering for charge carriers in the QW.

1. Introduction

Photoluminescence is an important tool which has been used extensively to study electronic behaviour in biased double-barrier resonant tunnelling (DBRT) structures. The techniques used may be classified into two broad categories according to the analysis used for extracting information from the PL data. In one case the energy, line shape and line width of features in the PL and photoluminescence excitation (PLE) spectra are of interest because they are directly influenced by the carrier densities in the quantum well (QW), through such mechanisms as phase space filling, renormalization and screening [1]. Thus, the analysis of these optical characteristics has given evidence for space-charge build-up in the QW on resonance, and the presence of a quantizing magnetic field has enabled determination of the carrier density from Landau level occupancy factors [2,3]. The second experimental category involves studying the intensity of the QW luminescence, as this is proportional to the product of the electron and hole densities in the QW and should therefore be capable of yielding additional information about the variation of electron density with bias. However,

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the interpretation of such data is complicated for the case of CW spectroscopy by the fact that it is not possible to carry out factorization of the observed variation of PL intensity with bias into a product of bias-dependent electron and hole densities. Thus the interpretation of PL intensity studies published to date has generally relied on assumptions about the QW hole population [4] and in the absence of detailed information on the minority carrier dynamics, such interpretations are susceptible to criticism [5]. Some progress has been made in the study of PL intensity by the use of time-resolved techniques [6, 7, 8]; Vodjdani *et al* [6] observed long-lived luminescence from a DBRT device illuminated by pulsed light below the QW absorption edge whilst Charbonneau *et al* [7] found that the PL rise-time differed according to whether the incident photon energy was above or below the QW absorption edge. Both observations indicate that carriers which are photo-excited in the contact regions may contribute to the QW PL emission. However, all previous time-resolved studies of DBRT devices have been incomplete in so far as they lack coverage over a range of biases and, as shown below, there is important structure in the bias dependence.

In the time-resolved PL spectroscopy reported here, information is obtained *directly* about the minority carrier dynamics from an analysis of the temporal evolution of the luminescence spectrum under conditions where the photo-injected electron represent only a negligible perturbation of the steady state population, which is due to tunnelling. Under these conditions the temporal evolution is completely dominated by the dynamics of the hole population. In order to form an unambiguous link between the observed temporal PL characteristics and a microscopic model of the DBRT device, information is needed on the time-integrated PL intensity as this gives an indication of how the radiative recombination probability for a hole in the QW varies with bias.

2. Experimental details

The sample used in this work is nominally identical to sample 2 of [3]; it is a $GaAs/Al_{0.33}Ga_{0.67}As$ DBRT structure grown by molecular beam epitaxy on an n-doped, (100)-oriented, GaAs substrate and etched to form 300 μm diameter mesas. Electrical contact was via the substrate and metallization on the top face of the mesa. Table 1 shows details of the MBE-grown layers and figure 1 shows schematically the conduction and valence band-edge profiles with an applied bias. The sample was held in an optical access cryostat and connected to an external bias source. A 30 Ω resistor was also connected via short leads to the device. The temperature of the sample was measured to be approximately 6 K during all of the experiments presented here. A current–voltage (I–V) characteristic is shown in figure 2 (solid line); note that there are two electron tunnelling resonances seen at 150 mV and 800 mV as expected for two quasi-cofined electron states in the well. The dashed line in figure 2 represents the I–V curve for the same device under intense pulsed illumination: 40 W cm^{-2} (CW equivalent). In terms of the perturbation of the I–V characteristic there was virtually no difference between the effect of pulsed and CW excitation provided the same average power level was maintained. Figure 2 shows that even under very bright illumination, which is over an order of magnitude higher than that used to collect the time-resolved PL data, the main effect of the laser light is to introduce a small shift in the positions of the tunnelling resonances—an observation which is significant in the interpretation presented later. A possible objection to these experiments could be that the bias measured externally does not reflect the band profile at the device as the transient, optically injected population undergoes charge separation and diffusion. The lifetime data presented below indicate, however, that in these experiments the optically injected carriers decay with a time constant which can be an appreciable fraction of the pulse repetition time. This in turn implies that the $I-V$ characteristic measured by the DC technique in fact gives a reliable guide to the nature of vertical transport even under pulsed illumination.

Table 1. Details of the layer structure in the DBRT device.

Layer	Doping	Thickness
n^+GaAs substrate	2×10^{17} cm^{-3}	
n^+GaAs	1×10^{18} cm^{-3}	0.5 μm
n^+GaAs	2×10^{17} cm^{-3}	0.5 μm
GaAs	Undoped	102 Å
$Al_{0.33}Ga_{0.67}As$	Undoped	85 Å
GaAs	Undoped	79 Å
$Al_{0.33}Ga_{0.67}As$	Undoped	85 Å
GaAs	Undoped	102 Å
n^+GaAs	2×10^{17} cm^{-3}	0.75 μm
n^+GaAs	1×10^{18} cm^{-3}	0.25 μm

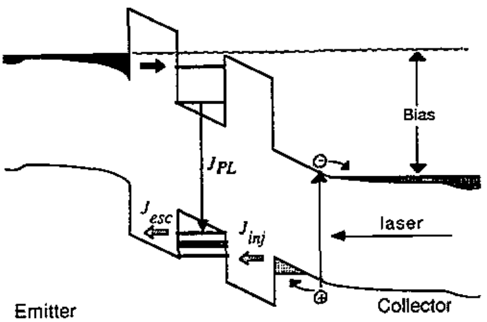

Figure 1. Spatial variation of band-edge energies in the vicinity of the DBRT structure at a bias near to the second electron resonance.

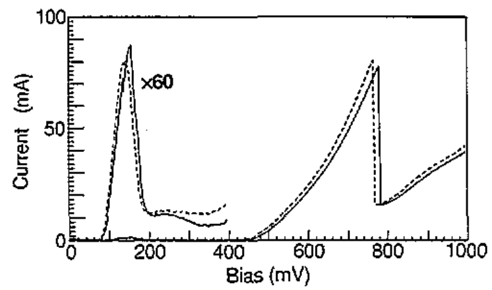

Figure 2. Current–voltage characteristic for the DBRT mesa held in helium gas at 6 K with a parallel stabilizing resistor connected. The resistor current has been subtracted from the data presented here. The solid line was recorded with the device in darkness; for the dashed line it was uniformly illuminated by light at 1.6 eV photon energy with a power density of 40 W cm^{-2}.

For time-resolved studies the optical excitation was provided by a synchronously pumped dye laser which delivered photons of energy 1.675 eV in pulses of several picoseconds duration which illuminated the mesa evenly. The luminescence emitted by the device was collected and dispersed by a 0.22 m spectrometer before passing into a streak camera arranged with the direction of the synchroscan time streak orthogonal to the monochromator dispersion. The resultant two-dimensional image was intensified by a micro-channel-plate intensifier then imaged onto a cooled silicon charge-coupled device (CCD). Figure 3 shows the intensity profile recorded by the CCD when the device was biased at 456 mV. This figure shows three PL peaks, which are in ascending energy: electron to acceptor recombination (bulk GaAs), bandgap recombination (bulk GaAs) and quantum well exciton recombination. Notice that the lifetime of the bulk bandgap PL is around 1 ns which is shorter than the QW luminescence. At short times after excitation the bulk GaAs carrier distribution is hot and this gives rise to a 'tail' in the bulk PL signal which extends to higher energy. This tail rapidly disappears as the carrier populations thermalize, and by 3 ns after excitation the bulk GaAs PL has become a single peak centred at 1.515 eV, the bulk GaAs free-exciton energy.

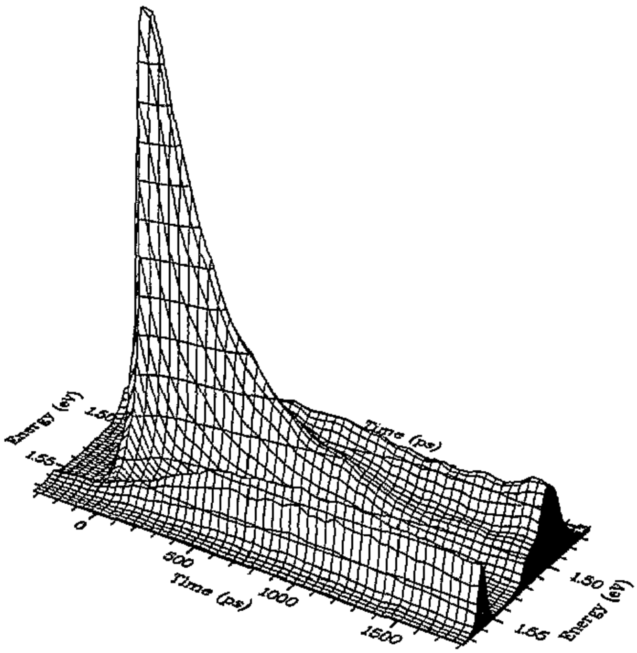

Figure 3. Time-resolved PL from the DBRT device biased at 465 mV, shown here as a contour plot on the energy–time plane.

3. Results and discussion

Figure 4 shows how the temporal evolution of the QW PL changes with bias. Only luminescence due to recombination between the lowest-energy confined states in the QW is shown because no higher-energy luminescence has been observed in this experiment (which would correspond to electrons in the second confined state), even when the device is biased at the second tunnelling resonance. This demonstrates that carriers tunnelling into the second confined level scatter rapidly into the lowest confined level, so that coherent tunnelling via the higher confined levels is unlikely as the carrier build-up associated with resonance will occur in the lowest levels: electron tunnelling is sequential, therefore. Notice that at low values of bias the lifetime of the signal is greater than the repetition time of our laser (13 ns). This gives rise to a non-zero PL signal prior to $t = 0$ which is due to PL emission from long-lived carriers injected by previous laser pulses [6]. Also, at short times the QW PL appears superimposed on the bulk GaAs hot carrier tail, the intensity of which has a complicated dependence on both energy and time, as can be seen in figure 3. Therefore, to produce the time-resolved PL curves shown in figure 4 we used an exponential fitting procedure to obtain an extrapolation of the energy dependence of the bulk PL through the region containing the QW PL for every time increment independently. We then subtracted our time/energy-dependent model of the bulk PL from the two-dimensional experimental data to reveal the true time evolution of the QW PL. In this way we were able to estimate the cooling rate for carriers in the bulk, as discussed later.

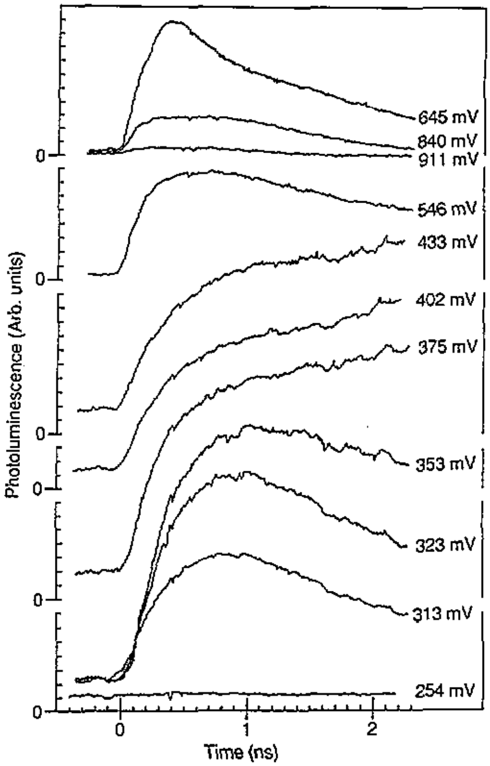

Figure 4. Time-resolved QW PL intensity following excitation by a picosecond laser pulse of photons at 1.675 eV. The sample was held in helium gas at 8 K.

The fact that the rising edge of the QW PL is clearly resolved implies that most of the PL is due to carriers which are photo-excited *outside* the QW and which then transfer into the well on a resolvable time scale, not by direct photo-excitation of carriers in the well. This strongly contrasts with the assumption by Young *et al* [4] that only direct excitation in the well contributes significantly to the production of PL. A number of authors [7, 9] have suggested that the difference may be due to different lifetimes for minority holes in the GaAs regions nearest the DBRT structure. Charbonneau *et al* [7] have demonstrated that even in devices containing no undoped spacer layer, a depletion region can be formed in the collector contact adjacent to the barrier which enhances the hole lifetime in this region and enables hole leakage into the QW to become significant.

Due to the high optical absorption of GaAs at 1.675 eV (around 2.7×10^4 cm^{-1} at 21 K [10]), virtually all the incident photons in each laser pulse will be absorbed in the top 1 μm of bulk GaAs. However, it has been shown [9] that subsequent luminescence which occurs at a lower energy than that of the laser photons (due to rapid thermalization of the photo-injected carriers) may penetrate the sample more deeply before being re-absorbed. This so-called *photon recycling* may extend a significant photo-injected population further into the structure, beyond the position of the barriers. The effect was shown to be an important route for photo-injected holes to reach the QW when the device is biased such that holes excited in the top contact drift away from the DBRT structure [9]. For the bias shown in figure 1 carriers injected by photon recycling beyond the barriers will not contribute to the hole population in the QW.

When photon recycling is taken into account a population of photo-injected holes is to be expected in the GaAs beneath the DBRT structure. However, the only mechanism which contributes to the hole population in the QW, *apart from tunnelling*, is direct absorption of the *original* laser photons, since the recycled photons have less energy than the fundamental absorption edge for the quasi-2D states in the QW and therefore cannot excite the inter-subband transition. When a bias voltage is applied to the structure the hole population in the (electron) collector region will be driven towards the DBRT structure and may accumulate in a layer adjacent to the collector barrier.

The accumulation of holes acts as a reservoir from which they tunnel into the QW, as indicted schematically in figure 1. Given that the electron–hole pairs injected optically have considerable excess energy, it is necessary to consider the possibility of hot carriers contributing to the flux entering the QW. This is found to be negligible however as the cooling time for carriers in the contact regions is several hundred picoseconds, as deduced from figure 3, whereas the QW PL continues rising towards a peak much later in time, so that most of the carriers in the contact regions will be cold when the QW PL peaks. Were the flux of hot holes significant this would lead to a peak in the QW PL at $t = 0$ followed by a decay on a timescale similar to the cooling time for the bulk; this is not observed, which implies that transfer of hot holes into the QW is insignificant.

The model outlined above must be invoked to account for the fact that the QW PL outlives the bulk GaAs PL and may even continue to rise after the bulk GaAs PL has decayed almost completely. Additional supporting evidence for this model is provided by the perturbation of the I–V characteristic: the shift to lower bias of the electron tunnelling resonances indicates that a larger field is present across the DBRT structure. This is consistent with a layer of positive charge accumulating adjacent to the collector barrier [6, 11]. The lifetime for holes stored in this layer will be very long in any DBRT device which has undoped buffer layers adjacent to the DBRT structure because the holes there are spatially separated from the electrons. The build-up of this population following arrival of the laser pulse depends on the depth profile of the injected hole density and will occur at different rates depending on the direction and magnitude of applied bias. According to a very simple diffusion model based on an estimated photo-injection profile, the hole density in the accumulation region reaches a maximum around 0.5 ns after the arrival of the laser pulse [12]. This reflects the fact that carriers which are photo-generated in the contact regions away from the DBRT add a significant weight to the hole accumulation at the barrier. Since the radiative lifetime for carriers which have accumulated in the undoped spacer layer is expected to be long, the principal mechanism by which they are removed is expected to be tunnelling through the collector, and this results in a population of holes in the QW. If the QW electron density remains constant in time then it is the temporal behaviour of this *tunnelling-supplied* QW hole population which determines the form of the curves shown in figure 4.

As the device bias is increased, four distinct regions of behaviour can be seen in the curves of figure 4: firstly, at biases below 290 mV there is no discernible step in the QW PL emission at $t = 0$, but its intensity varies from unobservable at zero bias to a finite level shown as the bottom trace in figure 4. Notice that this range of variation includes, at around 150 mV, the first electron tunnelling resonance, yet there is no observable temporal feature in the PL data which correlates with the expected variation of QW electron density as the bias increases through this value [13–15]. This observation contrasts with the behaviour reported in CW PL measurements [4, 9] by other workers who have seen the PL intensity correlate with the device current. Indeed, our measurement of time- and energy-integrated PL intensity (section 4) exhibits a similar correlation with device current at low injection levels.

Note that for a fixed photo-injected carrier density the integrated PL intensity measures the fraction of the photo-injected holes which recombine radiatively in the QW. Also note that below 290 mV bias the instantaneous PL intensity is never very high yet the integrated PL intensity can, for certain injection levels, attain its highest value—*due to the longevity of* *the PL emission.* In this light, an explanation for the low bias behaviour readily follows: at low bias, both electron and hole densities in the QW (n_e, n_h) are near zero and very little PL results. As the bias increases towards the first electron resonance n_e increases rapidly to a large value, giving a high quantum efficiency for radiative recombination of holes in the QW. Unless the carrier dynamics are dominated by non-radiative decay, this will ensure large integrated PL intensity. Noting the temporal behaviour of the PL emission, however, leads to the deduction that the probability of holes tunnelling into the QW from the collector contact must be small enough to limit n_h, despite the fact that the first heavy- and light-hole resonances should occur for a lower electric field than the first electron resonance [6, 16]. Thus at the first electron resonance, the low PL efficiency during the first 2 ns following excitation simply arises from a low hole population in the well.

With the bias between 290 mV and 360 mV the PL emission starts to rise sharply after arrival of the laser pulse and reaches a maximum within 0.5–1 ns, followed by a decay. The rise-time of the luminescence corresponds closely to the expected rise-time of the hole population in the accumulation layer due to the drift and diffusion of holes towards the barrier, so we assume that in this bias region hole transport into the QW from the accumulation layer is fast. The decay of the signal is associated with depletion of the hole accumulation in the undoped buffer layer. Notice that the rise of the PL signal remains dominated by the diffusive arrival of holes at all biases above this level, which indicates that the hole entry rate remains fast despite the increasing bias. It is possible that resonant tunnelling of holes into the QW could be responsible for this if the tunnelling proceeds via higher energy, lightly confined hole states, but this model predicts a gradual turning on of the PL rise as progressively higher energy hole states contribute to the flux of holes reaching the QW [6, 16, 17]. This is not the observed behaviour. To account for the sudden switching-on of the PL rise as the bias increases, possible models are that either holes in the accumulation layer begin to flow *over* the collection barrier as the bias increases beyond about 300 mV or (more reasonably) that hole tunnelling via some higher energy confined state(s) proceeds very much faster than tunnelling via the lowest energy heavy-hole-like state.

When biased between 370 mV and 450 mV the current through the device is at a local minimum in the I–V curve and so n_e is expected to be lower than at the biases previously discussed. This is reflected in the time-resolved PL curves: the QW emission still begins sharply after excitation but reaches a plateau or continues to rise slowly where previously the decay had set in, and does not reach the same peak intensity that it did before. This occurs because the lower value of n_e limits the radiative recombination rate for holes in the QW: hole tunnelling is fast whilst electron tunnelling is slow and the QW electron population becomes depleted. Under these conditions the tunnelling electron flux is more dominant in determining the PL level. This contrasts with the case where the optical injection level is reduced sufficiently to ensure that the photo-injected hole population arriving at the QW is smaller than the steady-state electron density. In this instance, saturation of the PL intensity is not expected and indeed at low enough injection levels is not observed, as shown in figure 5.

At biases greater than 450 mV we observe decreasing lifetimes for the QW PL emission until at around 910 mV PL is virtually absent, although the PL rise-time still indicates that holes can enter the QW freely. Further, the electron flux (as indicated by the device current) is around an order of magnitude higher than at some of the lower biases discussed above, which given the finite dwell-time for electrons in the QW [18], indicates that some electrons must be present in the well also. The shortening lifetime of the QW PL therefore indicates that the hole accumulation is emptying more rapidly after each laser pulse arrives and the

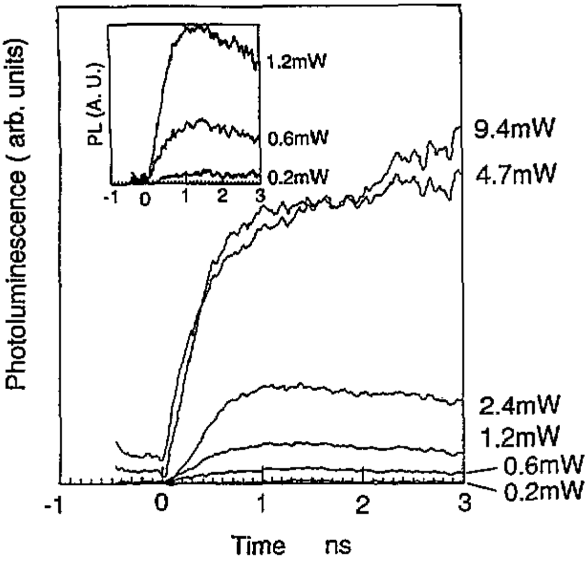

Figure 5. Variation of QW PL evolution with illumination intensity at a fixed bias level of 465 mV. Notice that saturation of the PL intensity is evident at the higher illumination levels but not at the lower ones.

dominant escape route for these holes is by passing into the QW. This is consistent with the assertion above that holes can enter the QW rapidly once the bias is above 360 mV. However the time-integrated PL intensity, which should reflect the balance between radiative and non-radiative channels for removal of holes, is smaller than it was at lower biases. To account for this we propose that the applied bias has shifted the valence band profile around the QW sufficiently to allow holes which pass over or through the first barrier to escape directly over the second barrier into the emitter contact.

4. Comparison with CW results

Only a portion of the photo-injected carriers due to each laser pulse ever recombine radiatively in the QW; the fraction which do so is related to the balance between radiative and non-radiative recombination channels for holes in the QW. Information about this balance can be obtained by measuring the time-integrated PL intensity. Our interpretation of the time-resolved PL results implies that distinct changes should occur in the time-integrated PL intensity as the device bias increases through the points which we identify with abrupt changes in the hole transport dynamics, namely the bias levels required to enable the accumulated hole population to flow rapidly past first the collector barrier and then the emitter barrier. This is indeed the case, as can be seen from figure 6 which shows the variation of the time- and energy-integrated PL intensity with bias for different optical power densities for both pulsed and CW excitation. These data were taken using a similar experimental configuration to the time-resolved experiments but with the streak camera replaced by a single photomultiplier. We have plotted the integrated luminescence intensity, rather than peak value because doing so reflects the balance between radiative and non-radiative recombination processes more accurately. This is especially true in the low bias region (below 200 mV) where both the PL lineshape and its position are strongly bias dependent, which is indicative of the complexity of the radiative recombination processes in the QW. In fact, there are at least two PL transitions separated by about 2 meV and the weight of the luminescence shifts abruptly between them at 100 mV of device bias. This phenomenon, the origin of which is not yet clear, is shown by the CW PL curves in figure 7.

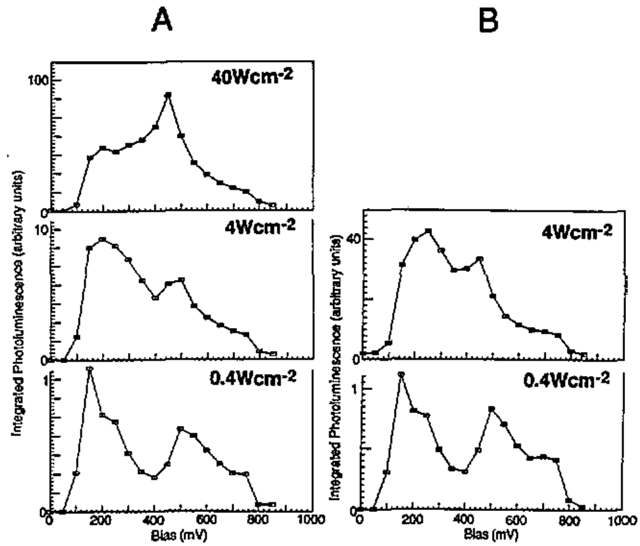

Figure 6. Time-integrated QW PL intensity recorded with (A) cw excitation, and (B) pulsed excitation. The photon energy in both cases was 1.6 eV and for the case of pulsed excitation the pulse length was several picoseconds. The power densities given correspond to the time-averaged optical power density.

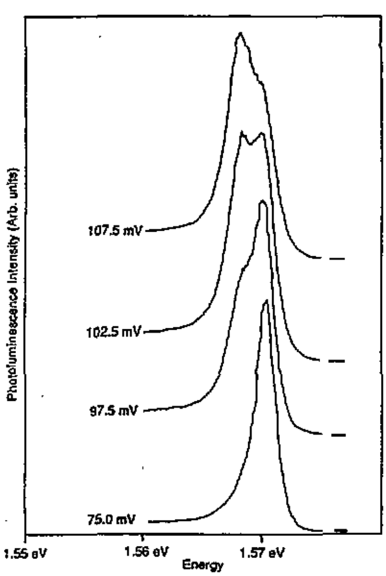

Figure 7. CW PL structure from the DBRT device around the 100 mV bias region, showing an unexplained shift in the weight of the PL transition between two closely separated components.

Note that the luminescence intensity exhibits a broad peak *between* the two electron resonances with a dip corresponding to depletion of the QW electron density at about 400 mV. The dip is most pronounced at low values of optical injection density. The increase in the hole entry rate is manifested by increasing luminescence from 100 mV bias onwards, but interpretation of this is confused by the fact that the QW electron density rises sharply in this region also. In contrast, at all biases greater than 500 mV the luminescence intensity decreases monotonically with increasing bias despite the fact that the QW electron density increases to a maximum at the second electron resonance, around 760 mV, therefore this provides a clear indication of the direct escape of holes from the QW, as discussed above.

5. Conclusion

We have studied the time-resolved photoluminescence arising from recombination of electrons and holes confined to the quantum well in a DBRT structure and inferred that photo-injected holes accumulate against the (electron) collector barrier, forming a 2D hole gas. The growth and decay of this population strongly affects the PL characteristics of our DBRT device which has an undoped buffer layer adjacent to the DBRT structure, since the lifetime of holes in this region is long enough to permit a significant population to accumulate and remain. When the population in this accumulation layer is allowed to become large enough holes may flow into the QW with a characteristic time which shortens drastically as the bias increases. The observed variation in the time-integrated PL intensity with bias is influenced by hole transport and reflects the balance between radiative and non-radiative recombination channels for holes. We have shown that the probability for holes escaping from the QW into the (electron) emitter contact becomes dominant at high biases and that inter-subband scattering occurs for carriers in the QW on a picosecond time-scale.

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