Thermally activated carrier escape mechanisms from $In_x Ga_{1-x} As/GaAs$ quantum wells

J. R. Botha and A. W. R. Leitch

Department of Physics, University of Port Elizabeth, P.O. Box 1600, Port Elizabeth 6000, Republic of South Africa

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The temperature dependence of the photoluminescence (PL) intensity of strained $In_xGa_{1-x}As/GaAs$ single-quantum-well (QW) structures grown by metalorganic vapor-phase epitaxy is investigated. We adopt the theoretical model recently proposed by Vening, Dunstan, and Homewood [Phys. Rev. B 48, 2412 (1993)] to describe the variation of the QW PL intensity with temperature. The Arrhenius behavior of the PL intensity at the highest temperatures investigated for each structure is shown to be due to the thermally activated escape of electron-hole pairs from the well. In each case, the deviation of the experimental data at intermediate temperatures is accounted for by assuming the dominant nonradiative recombination process to be the escape of the less-confined carrier species from the QW. The existing model is modified to include this process. The activation energies obtained are in reasonable agreement with the calculated heavy-hole confinement energies assuming a conduction-band offset ratio of $Q_c \approx 0.83$.

I. INTRODUCTION

With the current advanced state of epitaxial growth technology, the growth of quantum-well (QW) heterostructures has become almost routine. As a consequence, the dynamics of carriers in two-dimensional systems have been studied in considerable detail in recent years. However, no complete understanding of the carrier dynamics in QW's has been attained.

Nonradiative recombination significantly reduces the optical efficiency of a QW structure. The processes responsible for nonradiative recombination have not been uniquely identified. To date, mainly two mechanisms have been proposed by various groups, based on different experimental results. On the one hand, it has been suggested that recombination occurs through defects accumulated at or near the heterointerfaces.^{1,2} This process is usually described in terms of a surface/interface recombination velocity, although this concept should have minor relevance in QW's.³ For high-quality QW structures, on the other hand, it has been shown that the dominant nonradiative mechanism involves the thermally activated escape of carriers from the well, followed by recombination through defect states in the barrier material.⁴⁻⁹ This process has only recently been identified, on the basis of the Arrhenius behavior measured for the photoluminescence (PL) intensity^{4,5} and/or the Arrhenius behavior of the PL decay time.⁶⁻⁹ However, no consensus has been reached regarding the dominant carrier escape mechanism as the lattice temperature is raised. From studies of the temperature dependence of the PL intensity of $In_xGa_{1-x}As/GaAs$ QW structures, Lambkin et al.⁴ and Vening, Dunstan, and Homewood⁵ concluded that the escape of excitons or electron-hole pairs (bipolar emission) from the wells dominates at high temperatures ($\sim 50-300$ K). Bacher et al.^{6,7} reached the same conclusion from their investigations of the temperature variation of the PL decay time and PL intensity in $In_xGa_{1-x}As/GaAs$ QW's. For GaAs/Al_yGa_{1-y}As QW

structures, Gurioli et al.⁹ reported the main nonradiative mechanism to be the thermally activated escape of the less-confined carrier species (unipolar emission), based upon the activation energies obtained from the Arrhenius plot of the PL decay time. This group suggested that the Arrhenius plot of the PL intensity cannot be used to extract the activation energies of the nonradiative channels at elevated temperatures, due to the temperature dependence of the radiative recombination rate.

In this paper, the temperature dependence of the PL intensity of strained $In_x Ga_{1-x} As/GaAs$ QW's grown by metalorganic vapor-phase epitaxy (MOVPE) is reported. We adopt the recently proposed model of Vening, Dunstan, and Homewood⁵ (which includes the temperature variation of the radiative recombination rate) and show that the nonradiative mechanism at the highest temperatures is mainly related to bipolar emission from the QW's. Deviations of the data from the theoretical model at intermediate temperatures are suggested to be due to the emission of the less-confined species of carriers from the wells. The existing model has been adapted to account for this process. The activation energies obtained are in reasonable agreement with the calculated heavyhole confinement energies, assuming a previously reported value¹⁰ for the conduction-band offset ratio of $Q_c = 0.83.$

II. EXPERIMENTAL DETAILS

Four strained $In_xGa_{1-x}As/GaAs$ single QW structures were investigated. These structures were grown by atmospheric pressure MOVPE at 670 °C on undoped semi-insulating GaAs substrates [2° off (100) towards nearest (110)]. Details of the growth system have been presented elsewhere.¹¹ Trimethylgallium, trimethylindium, and 10% arsine in H₂ (all diluted in a palladium diffused H₂ carrier gas) were used as source materials. Each structure consisted of a single $In_xGa_{1-x}As$ layer sandwiched between a 6000-Å GaAs buffer layer and a

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1500-Å GaAs capping layer. The undoped material was nominally *n* type with 300-K free-carrier densities less than ~5×10¹⁴ cm⁻³. The nominal QW widths (L_z) were 25, 33, 58, and 100 Å. Layer thicknesses were determined from bulk growth rates (~8.3 Å s⁻¹ for $In_xGa_{1-x}As$; ~6 Å s⁻¹ for GaAs). The In mole fraction $(x = 0.192\pm 0.010)$ was deduced from x-ray diffraction measurements on ~3-4- μ m-thick calibration layers growth before and after the QW structures.

The PL response from these structures was studied between 12 and 240 K. The samples were mounted strainfree in a closed-cycle helium cryostat. The QW luminescence was excited indirectly using the 5145-Å line of an Ar-ion laser. The PL was analyzed with a 0.5-m spectrometer and detected with a photocathode having near S1 response. There can exist temperature variations between the temperature sensor and the sample. To circumvent this, each sample was mounted as close as possible to the sensor and the PL excited from a region less than 1.5 mm from the sensor. Sufficient time was allowed between scans to allow temperature stabilization. A laser power density of $\sim 30 \text{ W cm}^{-2}$ was employed, which corresponds to an excess carrier concentration in the wells of $\sim 10^{16}$ cm⁻³ at the lowest temperatures. This excitation density was sufficiently low to ensure no local heating of the sample¹² and to minimize band-filling effects. The QW luminescence were integrated numerically to account for the temperature-induced broadening of the peaks.

III. RESULTS AND DISCUSSION

The low-temperature PL response from all four structures is depicted in Fig. 1. Each spectrum comprises a single peak which is attributed to the recombination of excitons associated with the n = 1 electron and heavyhole subbands in the wells. The measured linewidths included in each spectrum indicate good heterointerface quality and compare favorably to some of the best values reported for similar MOVPE-grown structures.¹³ However, it may be emphasized that the linewidths are strongly influenced by the degree of substrate misorienta-



FIG. 1. Low-temperature PL spectra of four $In_{0.19}Ga_{0.81}As/GaAs$ single QW structures with nominal $L_z = 25$, 33, 58, and 100 Å. Laser power density ~ 30 W cm⁻². The baseline of each spectrum has been shifted for clarity.

tion.¹⁴ Linewidths well below 6 meV have been measured for similar structures grown in this laboratory on nominally (100) GaAs substrates.

The temperature dependence of the integrated PL intensity of each QW is depicted in an Arrhenius plot in Fig. 2. For all the samples the PL intensity is nearly constant at low temperatures. Above a characteristic temperature the intensity is reduced by orders of magnitude in each case. The onset of this reduction in intensity as well as the characteristic activation energy describing the intensity drop is clearly dependent on the well width L_z .

To quantify the quenching of the luminescence at higher temperatures, we adopt the model recently proposed by Vening, Dunstan, and Homewood⁵ as a first approximation. This group modeled the QW structure under indirect excitation by a simple rate-equation scheme, the principles of which will be repeated here in brief. Vening, Dunstan, and Homewood⁵ considered the multiple QW structure in which a single population n of photoexcited electron-hole pairs exists in the barriers and an independent population m_i in each of the w wells (i = 1to w). Using a constant excitation rate P into the barriers, trapping rate constants U_i into the wells, and detrapping rates $U_i\beta_i$ from each well, where

$$\boldsymbol{\beta}_{i} = \exp\left[-\frac{E_{A}^{i}}{kT}\right] \tag{1}$$

and E_A^i are the differences in energy between the barrier and QW luminescence, they assumed a nonradiative recombination rate constant R' in the barriers and a radi-



FIG. 2. (a) Arrhenius plots of the integrated PL intensity of the four strained $In_{0.19}Ga_{0.81}As/GaAs$ QW's with nominal $L_z = 25$ Å (i) 33 Å (ii) 58 Å (iii), and 100 Å (iv). Each set of data has been shifted vertically, for clarity. The dashed lines depict least-squares fits of Eq. (5) to the data, using $R \propto T^{-1}$; $U = R' \propto T$. In (b) the data are replotted for higher temperatures only, to illustrate the deviation from Eq. (5) in the region of the elbow.

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ative recombination rate constant R_i in each well. The rate equations describing such a system under steady-state conditions are

$$\frac{dn}{dt} = 0 = P - R'n - \sum_{i=1}^{w} U_i n + \sum_{i=1}^{w} U_i \beta_i m_i$$
(2)

and

$$\frac{dm_i}{dt} = 0 = U_i n - (U_i \beta_i + R_i) m_i . \qquad (3)$$

These two equations yield the following expression for the PL intensity from each well:

$$I_{i} = R_{i}m_{i} = P \frac{R_{i}}{\left[\beta_{i} + \frac{R_{i}}{U_{i}}\right] \left[R' + \sum_{j=1}^{w} \frac{R_{j}}{\beta_{j} + \frac{R_{j}}{U_{j}}}\right]}$$
(4)

In the case of a single QW, Eq. (4) reduces to

$$I = Rm = \frac{P}{1 + \frac{R'}{U} + \frac{R'}{R}\beta}$$
 (5)

This model inherently assumes that equilibrium between the carrier populations in the wells and barriers is established very quickly, which is not unrealistic since carrier capture and detrapping occurs on a time scale much faster than the recombination rates in the wells and barriers.¹⁵ Furthermore, the omission of several mechanisms, such as nonradiative recombination in the wells, radiative recombination in the barriers, and diffusion of photoexcited carriers to the QW's was assumed to have negligible influence on a fit to experiment. Vening, Dunstan, and Homewood⁵ showed that Eq. (4) adequately describes the Arrhenius behavior of the PL intensity of $In_xGa_{1-x}As/GaAs$ $In_xGa_{1-x}As/GaAs/$ and $Al_{\nu}Ga_{1-\nu}As$ multiple QW structures as well as carrier retrapping effects between wells.

Considering Eq. (5) for the single QW, it is clear that the temperature dependence is dominated by β . However, the characteristic temperature where strong PL quenching occurs, is strongly influenced by the value of R'/R. Hence, any temperature dependence of the rate constants is expected to influence the activation energies obtained from a fit to experiment. It has recently been shown that excitonic recombination dominates the PL of the single QW's under consideration up to the highest temperatures studied here.¹⁶ In the case of excitonic recombination in two dimensions, the recombination rate is expected from theory to vary as^{15,17}

$$\boldsymbol{R} \propto \boldsymbol{T}^{-1} \tag{6}$$

over a large range of the temperatures investigated in this work. In fact, Feldmann *et al.*¹⁸ and Akiyama *et al.*¹⁹ have measured the variation of the excitonic lifetime with temperature in GaAs/Al_xGa_{1-x}As QW's and found a linear increase up to ~50 K. We assume the relation in Eq. (6) to hold over the whole temperature range studied. It is less clear how R' and U vary with temperature. Following the arguments of Vening, Dunstan, and Homewood⁵ we assume both R' and U to increase linearly with temperature.

Returning now to Fig. 2, the dashed lines represent least-squares fits of Eq. (5) to the experimental data, employing the temperature dependences of the rate constants stated above. The best fitting parameters are listed in Table I. Also included are the best-fit parameters obtained by assuming different temperature dependences for the rate constants, to illustrate their influence on the activation energies E_A . The experimental activation energies $E_A(\exp)$ represent the difference between the GaAs band-gap energy²⁰ and the PL peak energy measured for each QW at high temperatures (T > 90 K) where exciton localization is negligible.¹⁶

Several features regarding the fitting parameters in Table I should be emphasized. Firstly, although the rate constants (R, R', U) are not uniquely determined, uniqueness of the relative values R'/U and R'/R is forced by the assumption R' = U. It should be noted, however, that this assumption does not significantly alter the activation energies E_A obtained from a least-squares fit to experiment. Secondly, it is not possible to determine the correct temperature behavior of the rate constants from the three sets of fitting conditions listed, since they fit the data equally well. However, only one set $(R \propto T^{-1})$; $U = R' \propto T$) predicts reasonable values for the rate constants. Assuming an excitonic recombination rate of 10⁹ s^{-1} at 12 K (corresponding to a low-temperature lifetime of ~1 ns²¹) this yields a value for $U \approx 4-5 \times 10^{13} \text{ s}^{-1}$ at 12 K. Although this is slightly higher than typical phonon frequencies ($\sim 8.8 \times 10^{12} \text{ s}^{-1}$ in GaAs), it agrees well with the values deduced by Vening, Dunstan, and Homewood⁵ for an In_{0.2}Ga_{0.8}As/GaAs multiple QW structure and with carrier trapping times of less than 1 ps measured by Oberli et $al.^{22}$ The other two sets of fitting conditions in Table I yield trapping rates U that are orders of magnitude higher than these values and therefore nonphysical. Furthermore, the assumption that all the rate constants are temperature independent yields activation energies consistently higher than $E_A(exp)$, in agreement with the results of Vening, Dunstan, and Homewood.⁵ Although the activation energies obtained by setting U=R'= const and $R \propto T^{-1}$ agree better with $E_A(\exp)$, the first set of fitting parameters $(R \propto T^{-1}; U = R' \propto T)$ seems to describe the physics of the process of carrier escape more accurately, based upon the more reasonable trapping rates predicted. The discrepancies between $E_A(exp)$ and the activation energies obtained from this set (for the three narrowest wells) can be accounted for by the experimental uncertainty ($\sim 5\%$). The large discrepancy for $L_z = 100$ Å is attributed to deviations of the temperature dependences of the rate constants from the simple relations assumed, at higher temperatures. It may be concluded from Table I that the dominant mechanism responsible for the Arrhenius behavior of the PL intensity at high temperatures is the thermally activated escape of electron-hole pairs from the QW's. This confirms previous experiments which showed that the thermal activation is determined by the total confinement energy of the electron-hole pair.⁴⁻⁷ The assumptions in-

TABLE I. Comparison of the activation energies E_A and relative rate constants U/R (at 12 K) obtained from a least-squares fit of Eq. (5) to the temperature variation of the integrated PL intensity of various $In_{0.19}Ga_{0.81}As/GaAs$ single QW structures, employing different temperature dependences for the rate constants R, R', and U. The values $E_A(exp)$ are the differences between the GaAs band-gap energy and the QW PL peak energy measured at high temperatures for each structure.

Nominal L _z (Å)	E _A (exp) (meV)	$R \propto T^{-1}$ $U = R' \propto T$		Fitting conditions $R \propto T^{-1}$ U = R' = const		R = const $U = R' = const$	
		E_A (meV)	U/R	E_A (meV)	U/R	E_A (meV)	U/R
25	74	68	3.7×10 ⁴	76	7.4×10^{5}	83	1.5×10^{7}
33	93	90	3.7×10^{4}	101	1.1×10^{6}	112	3.1×10^{7}
58	146	136	4.4×10^{4}	151	1.7×10^{6}	166	6.7×10^{7}
100	191	170	5.0×10 ⁴	187	2.2×10^{6}	204	1.0×10^{8}

herent to this rather simple model do not, however, allow one to determine from E_A whether electron-hole pairs or excitons are emitted into the barrier. Finally, it has been shown by Michler *et al.*⁸ that an activation energy equal to half the total confinement energy of the electron and hole may be expected in the low-injection case, where the residual doping level exceeds the photoexcited excess carrier density. The present results therefore confirm that our measurements were performed under high-injection levels.

Returning to Fig. 2 once again, it is clear that Eq. (5) does not simulate the elbow between the Arrhenius behavior at high temperatures and the saturated intensity at low temperatures. None of the fitting conditions attempted affects the sharpness of the elbow. Clearly, further physical effects should be taken into account. From a study of $In_x Ga_{1-x} As/Al_y Ga_{1-y} As$ single QW's, Vening, Dunstan, and Homewood⁵ suggested that one or several nonradiative mechanisms in the $Al_{\nu}Ga_{1-\nu}As$ barriers are responsible for the deviation of their data from Eq. (5) in the region of the elbow. However, a careful investigation of the data in Fig. 2 suggests an additional thermally activated process for each QW, characterized by an activation energy which increases with L_z . To account for the gradual decrease of the PL intensity at intermediate temperatures, it is assumed here that the dominating process is the emission of the less-confined species of carriers from the QW. In addition to the rate constants already defined, we assume a detrapping rate $U_1\beta_1$ for the less-confined carrier type from each well, where

$$\beta_1 = \exp\left[-\frac{E_1}{kT}\right] \,. \tag{7}$$

Here, E_1 denotes the confinement energy of the carrier and U_1 is the associated rate constant. The rate equations describing the single QW under steady state conditions are

$$\frac{dn}{dt} = 0 = P - R'n - Un + U\beta m \tag{8}$$

and

$$\frac{dm}{dt} = 0 = Un - (U\beta + U_1\beta_1 + R)m \quad . \tag{9}$$

All the symbols, except those defining the emission of the less-confined carrier species ($U_1\beta_1$), have the same meaning as in Eqs. (2) and (3), the subscripts *i* having been omitted for the single QW case. Note that the emission of one type of carrier from the well (unipolar emission) only enters in the rate equation describing the steady-state processes in the QW. Under high injection levels, the concentration of electron-hole pairs in the barriers is not expected to be influenced by the emission of the less-confined carrier type from the well. After solving for *n* from Eq. (8), substituting into Eq. (9) and rearranging, the PL intensity from the QW is given by

$$I = Rm = \frac{P}{\left[1 + \frac{R'}{U}\right] \left[1 + \frac{U_1}{R}\beta_1\right] + \frac{R'}{R}\beta}$$
(10)

The least-squares fits of Eq. (10) to the temperature dependence of the integrated PL intensity of the QW's are displayed in Fig. 3. The corresponding best-fit pa-



FIG. 3. Arrhenius plots of the integrated PL intensity of the In_{0.19}Ga_{0.81}As/GaAs single QW's with $L_z = 25$ Å (i), 33 Å, (ii), 58 Å (iii), and 100 Å (iv) for intermediate and high temperatures. The solid lines represent least-squares fits of Eq. (10) to the data assuming $U_1 = \text{const.}$

rameters and fitting conditions employed are summarized in Table II. It is evident that Eq. (10) simulates the data at intermediate temperatures satisfactorily. In describing the parameters and fitting conditions in Table II, the following argument should be mentioned: In order to explain the thermally activated escape of electron-hole pairs (bipolar emission), Michler et $al.^8$ and Bacher et $al.^7$ suggested that unipolar emission produces spatial charge accumulation. The resulting band bending, in principle, then lowers the barrier for carriers of opposite charge and leads to an enhanced emission of such carriers until equilibrium is reached. Therefore, in fitting Eq. (10) to the experimental data it is assumed in each case that the unipolar emission process only prevails at intermediate temperatures and that band bending does not sufficiently enhance the emission of the oppositely charged carriers in this temperature range. Thus, we have employed the activation energies E_A and relative rate constants U/Rlisted in Table I (for $R \propto T^{-1}$; $U = R' \propto T$) to describe the bipolar emission process at high temperatures. The activation energies E_1 describing the unipolar emission from the wells are listed in Table II. Also included are the expected heavy-hole confinement energies $E_{\rm hh}$ corresponding to a ratio of the conduction-band offset to the energy-gap discontinuity, $Q_c = 0.83 \pm 0.06$, reported by Andersson et al.¹⁰ The values of $E_{\rm hh}$ were calculated by a numerical solution of the Schrödinger equation for a finite square potential²³ [including the strain-induced band-gap increase²⁴ for $In_{0.19}Ga_{0.81}As$ and an exciton binding energy of 8 meV (Ref. 25)]. In the calculation we had to vary the nominal L_z by ~5-10% to match the total confinement energies $E_{A}(exp)$ measured for each well; obviously, different band offset ratios yield slightly different well widths.

As might be expected, the values of E_1 are very sensitive to the temperature variation of the detrapping rate constant U_1 , as illustrated in Table II. As was the case before, the two fitting conditions employed are indistinguishable in terms of the quality of the least-squares fits. The activation energies E_1 obtained by assuming U_1 to

TABLE II. Activation energies E_1 and relative rate constants U_1/R (at 12 K) obtained from least-squares fits of Eq. (10) to the temperature dependence of the PL intensity of the In_{0.19}Ga_{0.81}As/GaAs QW's. The activation energies E_A and relative rate constants U/R describing the Arrhenius behavior at high temperatures were taken from Table I ($R \propto T^{-1}$; $U=R' \propto T$). Also listed are the calculated heavy-hole confinement energies $E_{\rm hh}$ using $Q_c = 0.83 \pm 0.06$ reported in Ref. 10.

		Fitting conditions				
		$U_1 =$	const	$U_1 \propto T$		
Nominal L_z (Å)	$E_{\rm hh}$ (meV) ($Q_c = 0.83 \pm 0.06$)	E_1 (meV)	U_1/R	E_1 (meV)	U_1/R	
25	15±8	15	1.7	10	0.15	
33	18±9	24	6.4	18	0.41	
58	27±12	25	3.2	17	0.15	
100	33±13	31	2.7	22	0.11	

be constant, yield an average value of $Q_c = 0.83 \pm 0.06$, compared to a value of $Q_c = 0.87 \pm 0.06$ when setting $U_1 \propto T$. Both these results are in reasonable agreement with other reported values for the conduction-band offset ratio Q_c of around 0.8.^{10,26,27} It should be noted here that the conduction-band offset ratio Q_c in the strained $In_xGa_{1-x}As/GaAs$ system is still a controversial topic, despite numerous investigations. While the reported values for x < 0.3 range from $Q_c = 0.85$ to 0.4^{26} the majority of these values are concentrated in the range 0.6–0.8. The value of Q_c quoted in Table II has been chosen because the predicted heavy-hole confinement energies agree better with our E_1 values. For comparison, the opposite situation in which the electron is the lessconfined carrier type has also been considered. The expected electron confinement energies have been calculated for a range of values of Q_c , by following the procedure mentioned for the heavy hole. In this case, the activation energies E_1 correspond to $Q_c = 0.42 \pm 0.18$ (for $U_1 = \text{const}$) and $Q_c = 0.37 \pm 0.17$ (for $U_1 \propto T$). These values are significantly lower than the prevailing opinion. Also, for a specific fitting condition, there is a large scattering between the values of Q_c predicted by the activation energies obtained for different QW's. Furthermore, in order to match the activation energy obtained for each QW to an expected electron confinement energy, the nominal L_z had to be varied by ~25-40 %, which is larger than the uncertainty in the growth rate $(\sim 10-15 \%)$. Hence, it is believed that the heavy hole is the less-confined species of carriers in this QW system.

Finally, it is significant to note that both fitting conditions in Table II predict values for the relative rate constant U_1/R that are orders of magnitude lower than the values for U/R deduced earlier. In fact, the U_1 values are comparable to the low-temperature radiative recombination rate constant R in the QW's. These much lower rates predicted for the unipolar emission process are not surprising. It is suggested that the emission process is limited by the rate of exciton scattering events (which lead to a dissociation of excitons), as well as the probability of the electron being recaptured by a hole to form an exciton. The latter factor would account for the competition between carrier escape and the reformation of an exciton after a scattering event. As the temperature is raised, the resultant influence of these factors should lead to an increase of the emission rate constant. However, without knowledge of the correct temperature behavior of these limiting factors, this point will not be pursued any further. It should only be stressed that a temperature dependence for U_1 of the form $U_1 \propto T^p$ (p > 1) yields values for U_1 so much lower than those expected from the low-temperature escape time (20-30 ps) measured for GaAs/Al_vGa_{1-x}As shallow QW's (Ref. 28) as to render them physically meaningless.

IV. SUMMARY

The temperature dependence of the PL intensity of four strained $In_{0.19}Ga_{0.81}As/GaAs$ single QW structures grown by metalorganic vapor-phase epitaxy, has been investigated. To quantify the experimental data, we have

adopted a recently proposed model⁵ which takes into account the expected temperature variation of the rate constants describing the steady-state processes in the structures. The Arrhenius behavior of the QW PL intensity at the highest temperatures studied for each structure was shown to be due to bipolar emission from the QW. The existing model has been modified to improve the description of the PL quenching at intermediate temperatures, which was suggested to be related to the thermally activated escape of the less-confined carrier species (unipolar emission). For each QW, the theoretical expression derived excellently simulates the variation of the PL intensity over the whole range of temperatures and yields physically reasonable parameter values. Assuming the less-confined carrier type to be the heavy hole, we deduced a conduction-band offset ratio Q_c between 0.83 and 0.87.

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