Dependence of the Electroluminescence on the Spacer Layer Growth Temperature of Multilayer Quantum-Dot Laser Structures

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***Abstract*—Electroluminescence (EL) measurements have been performed on a set of In(Ga)As–GaAs quantum-dot (QD) structures with varying spacer layer growth temperature. At room temperature and low injection current, a superlinear dependence of the integrated EL intensity (IEL) on the injection current is observed. This superlinearity decreases as the spacer layer growth temperature increases and is attributed to a reduction in the amount of nonradiative recombination. Temperature-dependent IEL measurements show a reduction of the IEL with increasing temperature. Two thermally activated quenching processes, with activation energies of ~ 157 meV and ~ 320 meV, are deduced and these are attributed to the loss of electrons and holes from the QD ground state to the GaAs barriers. Our results demonstrate that growing the GaAs barriers at higher temperatures improves their quality, thereby increasing the radiative efficiency of the QD emission.**

***Index Terms*—Activation energy, electroluminescence (EL), quantum dot (QD), spacer growth temperature.**

I. Introduction

SELF-ASSEMBLED quantum dots (QDs) have attracted considerable interest in recent years for their use in lasers due to the prediction that the threshold current density will be lower and less sensitive to temperature, compared to conventional quantum well (QW) or bulk devices [1]. Most recent work in this area has involved the growth of InAs–GaAs dots in an InGaAs well—the so-called dot-in-a-well (DWELL) structure-to achieve 1.3 μm emission at room temperature (RT). This approach also increases the quantum dot (QD) density with respect to the InAs QDs grown in GaAs and helps the QDs to capture carriers more efficiently [2], [3]. Although significant improvements have occurred in QD laser performance in recent years, some of the initial theoretical predictions such as temperature insensitive operation have yet to be achieved in practice, possibly in part due to nonoptimized growth conditions. Most practical laser structures require multiple stacks of QD layers to increase the overall QD number, and hence provide sufficient gain for the laser. The GaAs spacer layers that separate the QD stacks are required to provide a smooth growth surface for subsequent dot layers so that the characteristics of each layer are identical. Any roughness of this spacer layer can lead to degradation of subsequent QDs grown [2]. In addition multiple stacks of QDs can result in the accumulation of strain as subsequent layers are grown [3] and therefore growth conditions have to be optimized to avoid defects and surface roughness from occurring.

One popular approach to optimize QD structures is by thermal annealing, which appears to improve the quality of the QD laser structures [5]–[8]. Post growth annealing has been shown to remove large dislocated dots [4] and reduce the dislocation density [5]. More recent work has concentrated on *in situ* annealing of the spacer layer, which also appears to improve the performance of the QD lasers [4], [8]. The spacer layer in almost all InAs QD lasers is GaAs and it is well established that growing GaAs by molecular beam epitaxy (MBE) at high temperatures improves its carrier lifetime and decreases the defect concentration [6]. A further consequence of high temperature growth of GaAs has been reported to be an increase in the smoothness of the GaAs–InGaAs interfaces [7]. However, there is a tradeoff in growing the GaAs spacer layers at the highest possible temperature to improve the optical quality and the risk of evaporating indium from the dots, which results in a blue shift of the emission. Liu *et al.* [8] have shown recently that for structures with 50 nm GaAs spacer layers, growing the initial 15nm of the spacer layer at a relatively low temperature of 510 °C (the same temperature used to grow the quantum dots) and then increasing the growth temperature to 580 °C for the remaining 35 nm improves the lasing performance. This improvement has been attributed to the removal of large defective dots which occur when the entire spacer layer was grown at 510 °C, as shown by transmission electron microscopy (TEM) [8]. It was suggested that growing the entire spacer layer at 510 °C provides a rough growth surface, with QDs nucleating preferentially in pits on the surface and subsequently developing into large defective dots. Increasing the growth temperature for the latter part of the GaAs resulted in a much smoother surface. Further increases in the growth

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temperature of the GaAs spacer layer beyond 580 °C showed further improvements in the laser device performance [9], even though there was little indication of the presence of large defective QDs in the cross-section TEM of samples grown at 580 °C. The reason for the continuing improvement in the modal gain is not clear.

To investigate the dependence on the growth of the GaAs spacer layer in more detail, we have undertaken a study of the injection current dependence of the electroluminescence (EL) from a set of three InAs DWELL laser structures with different GaAs spacer layer growth temperatures. These samples have significantly different lasing characteristics, despite having some common properties (such as the composition and basic electronic structure) suggesting that they have significantly different material quality.

II. Experimental Details

A. Sample Preparation and Fabrication

All samples were grown by solid source MBE on silicon doped GaAs substrates as detailed in [11]. Briefly, the active region of the laser structures comprised of five layers of InAs QDs separated by 35-nm GaAs spacer layers. Each dot layer consisted of 3.0 monolayers (MLs) of InAs grown on 2-nm $In_{0.15}Ga_{0.85}As$ and covered by 6-nm $In_{0.15}Ga_{0.85}As$ to give a DWELL structure. The active region was surrounded by undoped GaAs, with n- and p-type AlGaAs layers completing the waveguide structure. Full growth details are given in reference [11]. A p+ GaAs layer was grown on top of the upper p-type AlGaAs cladding layer to enable low resistance ohmic contact to be formed. Growth temperatures were 620 °C for the AlGaAs and 510 °C for the In containing layers. In this series, the temperature at which the GaAs spacer layers were grown was varied. All 35 nm of the GaAs spacer layer for sample A was grown at a substrate temperature of 510 °C, i.e., at the same temperature as the In containing layers. For samples B and C, the GaAs spacer layer growth sequence was divided into two parts, with the initial 15 nm thickness grown at 510 °C and the remaining 20 nm grown at higher temperatures of 580 °C and 620 °C, respectively. It was found that sample A did not lase with continuous-wave (CW) current at RT, sample B showed only excited state lasing, while sample C gave the best performance with ground state CW lasing and a low threshold current density [9].

The optical properties of QD structures are often investigated by photoluminescence (PL) [13]–[15], however, this is often difficult to measure in full laser structures without etching off the contacting layers or relying on investigations of simpler test structures, which may not be representative of the full device structure. In PL measurements, variations in the thickness and doping levels of the top cladding layers can also affect the injection of minority carriers into the structure. In contrast, the injection of carriers into the active region of laser structures in EL measurements can be controlled very accurately.

From the three grown samples, circular mesa diodes with diameters of 50 μm, 100 μm, 200 μm, and 400 μm were fabricated by optical lithography and wet chemical etching. Top annular contacts were deposited to enable the EL to be extracted from the top surface. This, together with the circular shaped mesas avoids the effect of amplified spontaneous emission. Mesa diodes from each sample were probed at RT and dark current-voltage (*I-V*) characteristics were obtained. Both forward and reverse currents for different sized diodes were found to scale with area, suggesting that all the current was due to bulk mechanisms and that the edge leakage currents were negligible.

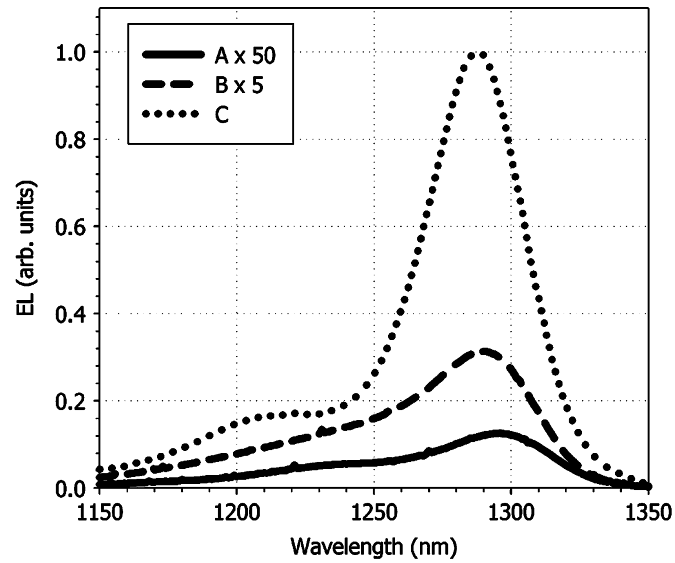

Fig. 1. RT EL spectra of samples A, B, and C with 1-mA injection current plotted normalized to sample C.

The EL spectra were recorded under constant current conditions on 400 μm diameter devices bonded on TO5 headers. Light emission from the surface of the mesa diodes was collected and mechanically chopped, before being spectrally dispersed and detected by a liquid nitrogen cooled germanium detector using standard lock-in techniques. The position of the devices was carefully optimized before the measurements. The devices were placed in a closed cycle helium cryostat to enable EL measurements from RT to 10 K, a heater stage was used for measurements from RT up to 450 K.

III. Experimental Results

Fig. 1 shows the RT EL spectra for the three samples for a constant injection current of 1 mA where the plots are normalized to that of sample C. It is clear that the EL intensity increases with increasing spacer growth temperature, with sample C giving almost two orders of magnitude higher EL intensity compared to sample A. There is a slight blueshift in the ground state emission wavelength as the spacer growth temperature is increased, corresponding to a shift of 6 meV between samples A and C as detailed in Table I. This slight blueshift might be due to a small loss of indium from the QDs as the growth temperature of the upper part of the GaAs spacer layer is increased. The shorter wavelength peaks that can be clearly seen in all three samples are believed to be the first excited states.

EL spectra measured at different injection currents were then integrated to qualitatively measure the total amount of luminescence collected. Fig. 2 shows the integrated EL intensity (IEL)

TABLE I

VALUES FOR THE RT EL PEAKS AND THE QW/WL PHOTOCURRENT PEAKS FOR SAMPLES A, B, AND C. ALSO GIVEN ARE THE EXPERIMENTALLY DETERMINED ACTIVATION ENERGIES FOR THESE SAMPLES AND THE SEPARATION BETWEEN THE VARIOUS ENERGY LEVELS. QD_1 AND QW_1 REFER TO THE GROUND STATE OF THE QD AND INGAAS QW/WL RESPECTIVELY

	Peak QW_1/ WL (PC) (eV)	Peak QD_1 (EL) (eV)	QW_1 /WL-GaAs (meV)	QD_1 – QW_1/WL (meV)	QD_1-GaAs (meV)	E_{a1} (meV)	E_{a2} (meV)	2 : 1 ratio of QD_1-GaAs (meV)
A	1.286	0.957	134	329	463	330	157	308 : 154
B	1.265	0.961	155	304	459	316	157	306 : 153
C	1.265	0.963	155	302	457	311	153	304 : 152

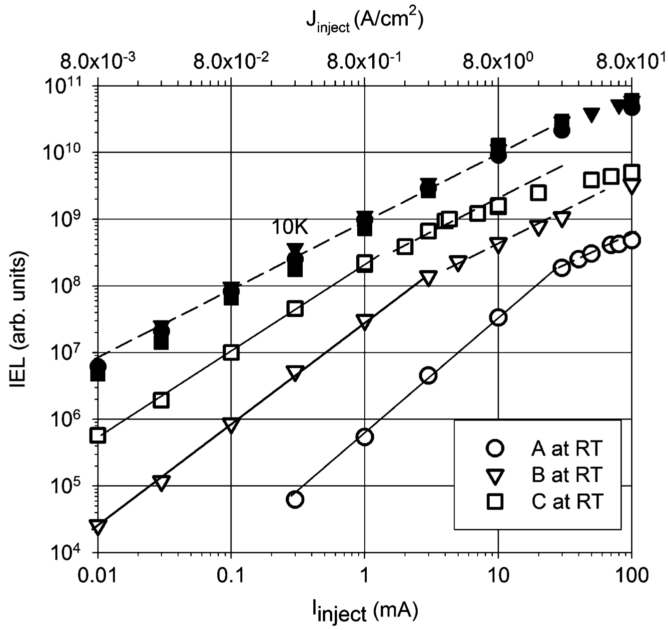

Fig. 2. IEL versus I_{inject} and the equivalent current density J for samples A, B, and C at RT (open symbols) and 10 K (closed symbols). Fittings for region I (solid lines) and II (dashed lines) are also shown.

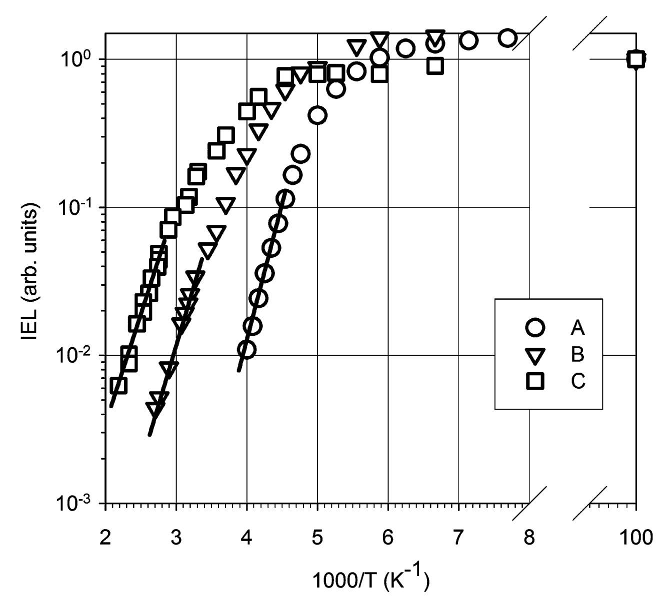

Fig. 3. Arrhenius plots of the temperature dependence of the IEL of samples A, B, and C.

versus injected current, I_{inject}, over the range from 0.01 to 100 mA on a log-log plot at both RT and 10 K. At 10 K, all three samples have a linear dependence of IEL on I_{inject} (a gradient of 1 in the log-log plot) with a similar magnitude of IEL across a large range of injection currents. In contrast, at RT there is a significant difference in the magnitude of the IEL between the samples, especially at lower injection currents. For each sample, two characteristic regions can readily be identified. Region I shows a superlinear dependence of IEL on I_{inject} (gradient larger than 1 in the log-log plot) and occurs at low injection currents. Region II occurs at an intermediate injection current and consists of a small region where there is a linear dependence of IEL on I_{inject} (gradient becomes approximately 1 in the log-log plot). Finally, at high injection currents there is a third region, seen most clearly in sample C, where the IEL increases sublinearly with I_{inject}.

The gradients in region I decrease as the spacer layer growth temperature increases, from 1.7, 1.5, to 1.3 (in the log-log plot) for samples A, B, and C, respectively. The region I/II boundary also appears to occur at lower currents as the spacer layer growth temperature increases. Region II is clearly visible for sample A and B, but is less obvious in sample C, while region III is only obvious for sample C over the injection current range studied.

Measurements were undertaken to observe the changes in the IEL as the temperature is varied from 10 to 450 K. A plot of ln(IEL) against the inverse of temperature to obtain the Arrhenius plot is shown in Fig. 3. A constant injection current of 0.1 mA (0.08 A/cm^2) was used throughout these measurements as it gave sufficient luminescence for the worst sample, sample A, but avoided high injection conditions where Auger effects may be significant. The shape of the plots in Fig. 3 is similar to the temperature dependence of the integrated QD photoluminescence [10]–[12]. The plots consist of three regions, a low temperature region, an intermediate temperature region and a high temperature region. At low temperatures, the IEL intensity is not temperature dependent and has a similar magnitude for all three samples, consistent with the data in Fig. 2. At intermediate temperatures, the IEL intensity appears to decrease slowly with temperature. At high temperatures, the IEL decreases exponentially with temperatures and an activation energy, E_{a1}, can be determined for each sample as shown in Table I. The temperature beyond which the IEL intensity starts to decrease, increases with the GaAs spacer growth temperature. However, the activation energy deduced for all samples is approximately 320 meV.

IV. DISCUSSION

The dependence of the IEL intensity on I_{inject} can be described by a simple analysis of the rate equation

$$\frac{dn}{dt} = G - \frac{n}{\tau_{\rm nr}} - Bnp \quad (1)$$

where G is the carrier generation rate, $\tau_{\rm nr}$ is the nonradiative recombination lifetime, B is the radiative recombination coefficient, n and p are the electron and hole concentrations, with Bnp the radiative recombination. At steady state, $dn/dt = 0$, and since the generation of the carriers is through the injection of current, $I_{\rm inject} \propto G$.

When radiative recombination is dominant, (1) becomes $Bnp \propto I_{\rm inject}$, giving a linear dependence of Bnp on $I_{\rm inject}$ [10]. When nonradiative recombination is dominant, $Bnp = BG^2\tau_{\rm nr}^2$, which gives $Bnp \propto I_{\rm inject}^2$ and a square law dependence of the IEL intensity on $I_{\rm inject}$.

In Fig. 2, at low injection currents (region I) the power law coefficient of the superlinear dependence of IEL intensity on $I_{\rm inject}$ decreases from sample A to sample C, indicating that the relative amount of nonradiative recombination in the samples is reduced as the spacer growth temperature increases. The number of carriers needed to saturate the defects responsible for the nonradiative recombination may also be expected to vary, this would explain why sample A requires almost 30 mA before the gradient becomes 1 whereas sample C requires a current that is lower by more than an order of magnitude.

Fig. 4 shows the linear region of Fig. 2 for all three samples, plotted on a linear-linear graph. At 10 K, the differences in the gradient and the magnitude of the IEL between the samples are not significant. However at RT, it is clear that the radiative efficiency, defined as the ratio of the amount of light collected (IEL) to the carriers injected $(I_{\rm inject})$, for the samples, increases with the spacer layer growth temperature. From Fig. 2, there is a clear decrease in the gradient at high current injection levels for sample C. Identical behavior was obtained using a pulsed current source, ruling out heating effects. It is possible that other nonradiative recombination processes such as Auger recombination [13], [14] might begin to become significant at these high current levels.

Comparing the superlinear regions of the three samples at RT (region I), we find that the magnitudes of the IEL for a given injection current differ. At 10 K however, all the samples have a similar magnitude of IEL and a gradient of 1 even at very low injection currents. This may be due to all the defects being filled and inactive, or that there is minimal carrier escape from the dots at very low temperatures. By comparing the 10 K and RT IEL at 3 mA injection current, it can be seen that even the highest spacer layer growth temperature, sample C, is still only $\sim 20\%$ efficient at RT, assuming an efficiency of 100% at 10 K. Sandall *et al.* reached a similar conclusion for a slightly different In(Ga)As DWELL structure grown in the same reactor to the present devices but with 50 nm GaAs spacer layers, for which the first 15 nm was grown at 510 °C and the latter 35 nm at 580 °C. By determining the nonradiative recombination component of the injected current in absolute units a room temperature radiative efficiency of 20% at threshold was deduced. [15]

A linear dependence of the IEL with injection current has previously been shown to imply low defect and dislocation densities [16], [17]. Early work by Ding *et al.* [18] on multiple

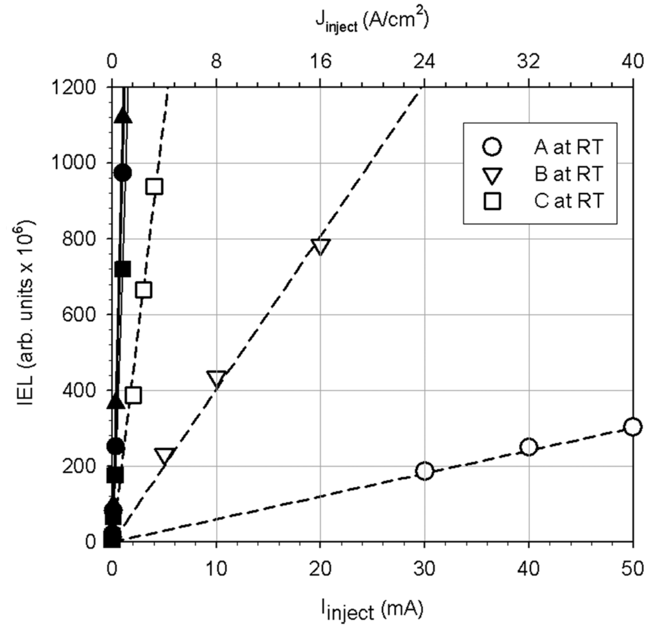

Fig. 4. IEL versus $I_{\rm inject}$ and the equivalent current density J for samples A (◯), B (▽) and C (□) at RT (open symbols) and 10 K (closed symbols).

quantum wells attributed the transition from a quadratic to a linear dependence of the PL intensity on the laser excitation power to the competition between nonradiative recombination at nearly saturated interface traps and radiative recombination. Recently, Sanguinetti *et al.* [10] observed a superlinear dependence of the integrated PL on laser excitation power for a QD structure and suggested that this behavior requires the presence of efficient nonradiative channels. Le Ru *et al.* [12] also reported on a superlinearity in the PL measurements of annealed QD structures, however they attributed this to the effect of the capture by the QDs of uncorrelated pairs of electrons and holes. We believe that in our samples the superlinearity observed is due to the presence of traps or defects in the samples, given that the samples are identical except for the spacer layer growth temperature.

The reduction in the IEL intensity with increasing temperature above a certain critical temperature, as observed in Fig. 3, implies a temperature activated quenching mechanism. The activation energies obtained for samples A, B and C can be attributed to the escape of carriers from the QDs to the barrier or wetting layer [12], the effect of temperature activated traps or a combination of both processes. The similar activation energies for all the samples are not unexpected since the structures are nominally identical. The measured energy separation between the QD ground state emission and the GaAs barrier (GaAs-ΔQD$_1$), shown in Table I for all three samples is $\sim$460 meV. The deduced activation energies of 311–330 meV could therefore correspond to electron escape from the QD ground state to the GaAs barriers if a 2:1 ratio in the values of the effective barrier height for electrons to holes is assumed. However, these activation energies only fit the IEL behavior at high temperatures of > 240 K, > 280 K, and > 320 K for samples A, B, and C, respectively. This suggests the possibility of a second activation energy at intermediate temperatures.

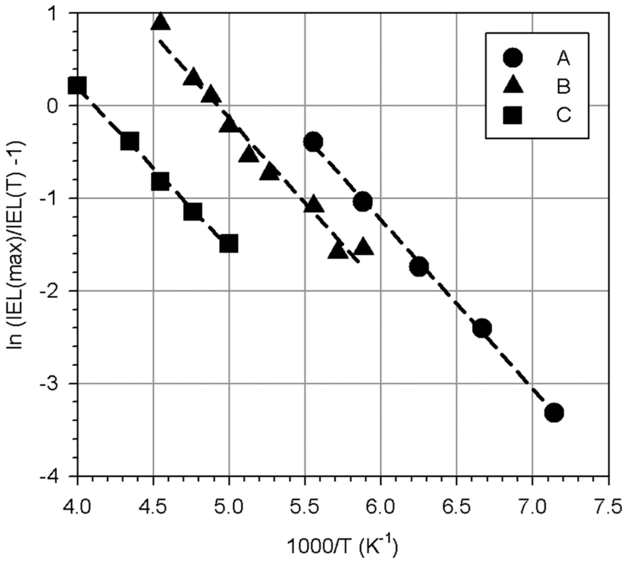

Fig. 5. IEL temperature dependence versus inverse temperature presented as {IEL(max)/IEL(T)-1} versus 1/T over an intermediate temperature range for samples A, B, and C.

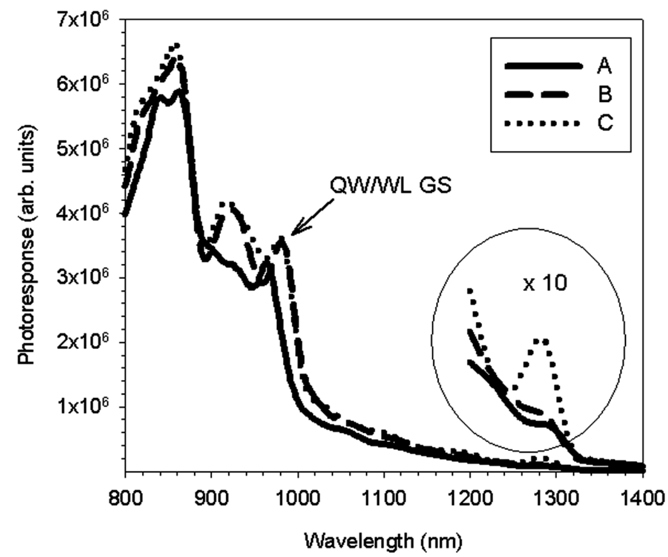

Fig. 6. RT photocurrent spectra for samples A, B, and C. Inset shows the spectra in the region of the QD ground state magnified by 10 times.

Torchynska *et al.* [19] have shown that by plotting ln[IEL(max)/IEL(T)−1] as a function of inverse temperature over this intermediate temperature range, a straight line relationship is obtained, where the gradient corresponds to a second activation energy. IEL(max) is the maximum intensity of the IEL obtained during the experiment while IEL(T) is its value at temperature, T. Plotting the data in this way allows the intermediate temperature behavior to be more easily seen, permitting an activation energy to be more accurately determined. Fig. 5 plots the data in this form and reveals that a straight line behavior is observed over this intermediate temperature range for all three samples. The gradient in this region corresponds to an activation energy, Ea_2, as detailed in Table I with values between 153–157 meV for all three samples. It is interesting to note that the sums of the two activation energies (311–330 meV and 153–157 meV) is very close to the measured value of QD_1-GaAs. This results suggests that as the temperature increases holes may initially be lost from the QDs followed by the loss of electrons at higher temperatures.

The present activation energies are significantly different to the values obtained by Torchynska *et al.* [19] of 370 meV and 80 meV on a three-stack InAs QD in $In_{0.15}Ga_{0.85}As$ QW structures with 30 nm spacer layer thickness. The 370 meV value was attributed to exciton escape from the QD ground state to the InGaAs QW, while the 80 meV value was attributed to exciton loss from high energy excited states related to the InGaAs QW. To investigate the possibility that the activation energies obtained in samples A, B and C are also due to similar loss processes via the InGaAs QW/wetting layer, room temperature photocurrent (PC) measurements were undertaken and the results are shown in Fig. 6. These show clearly the presence of peaks at 1.286 eV for sample A and 1.265 eV for samples B and C. These peaks can be attributed to the InAs wetting layer (WL) or InGaAs QW ground state transition. Hence the initial activation energy Ea_2 of $\sim$ 157 meV could explain the loss of excitons from this WL/QW transition to the GaAs barrier, given by QW_1-GaAs in Table I, for samples B and C, but not for sample A where the separation of 134 meV is significantly less than this measured activation energy. Hence, it seems more probable that this $\sim$ 157 meV activation energy is due to the loss of holes from the QD ground state to defects in the GaAs barrier for all the samples, followed then by the loss of electrons with a $\sim$ 320 meV activation energy at higher temperatures.

There have been several reports recently on the effects of p-type modulation doping on the performance of QD laser structures. The p-doping appears to give a more temperature independent laser threshold current density $(J_{\rm th})$ around room temperature but at the expense of higher values of $J_{\rm th}$. The p-doping is thought to increase the confinement of electrons in the dots via their Coulombic attraction to the extrinsic holes. To date there have been no reports of measurements reported in this paper applied to p-doped structures.

At present, it is unclear why Torchynska *et al.* obtained such different activation energies for nominally similar structures. Nevertheless, regardless of the exact details of the escape mechanism, the fact that the RT IEL intensity is a sensitive function of the GaAs growth temperature implies that the optical properties are sensitive to the quality of the GaAs and that a high growth temperature for the spacer layer is always desirable. Although the large dislocated dots reported by Liu *et al.* [8], which occur when a low GaAs growth temperature is used, may also contribute to the reduction of the radiative efficiency, the present results suggest that nonradiative recombination in the GaAs also contributes reducing these processes may offer the possibility of higher performance QD lasers.

V. Conclusion

We have studied the IEL as a function of both injection current and temperature for a series of nominally identical QD laser structures but with different spacer layer growth temperatures. The relative IEL intensities and the dependence of IEL on injection current and temperature suggest that a higher spacer layer growth temperature reduces the nonradiative recombination in the structure. From the temperature dependence of the IEL, two activation energies of ~ 157 meV (observed over the temperature range of 140 K to 250 K) and ~ 320 meV (observed at higher temperatures) are extracted, which are almost identical for all three samples. There are two possible mechanisms that may account for these activation energies. The first involves the loss of holes from the QDs to the GaAs barrier followed by the loss of electrons at higher temperatures. The second involves the loss of excitons from the InGaAs WL/QW to the GaAs barrier, followed by the escape of excitons from the QDs to the InGaAs WL/QW. We believe that the first mechanism provides better agreement with our experimental data. Irrespective of which mechanism dominates, it appears that improving the quality of the GaAs barrier can reduce the nonradiative processes in the structure.

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