Dual-state lasing and the case against the phonon bottleneck

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ABSTRACT

Quantum Dot lasers exhibit the novel phenomenon of dual state lasing where population inversion can be achieved on two optical transitions within the dots. In principle this might occur if a phonon bottleneck exists to impede relaxation of carriers from the higher energy state. Here we present an alternative explanation whereby different lasing modes compete for carriers and are spatially separable. Evidence comes from a comparison of electrical and optical measurements made on the devices. The evolution of a particular lasing mode depends on diffusion of carriers between dots and we show how, using an equivalent circuit model, this is consistent with our measurements.

Keywords: Quantum dots, III-V semiconductors, carrier dynamics, dual state lasing

1. INTRODUCTION

InAs/GaAs quantum dot (QD) lasers have been the focus of intensive research efforts because they offer the possibility of optoelectronic sources in the 1000 – 1500+ nm wavelength range, grown on GaAs substrates [1, 2], and because the three-dimensional confinement of electrons and holes in the QDs gives rise to discrete electronic states, which are expected to result in low threshold currents [3] and temperature insensitive operation [4]. However, the overall emission from the ensemble of QDs is inhomogeneously broadened by variations in QD size and composition and the operation of a QD laser will be determined by the collective behavior of the QD ensemble, resulting in novel phenomena such as dual-state lasing, where simultaneous lasing involving both the ground state (GS) and first excited-state (X1) transitions is observed [5-7].

Dual-state lasing has previously been attributed to a bottleneck effect which limits the speed of carrier relaxation between X1 and the GS of the QDs leading to a build-up of carriers in the X1 level [5]. At threshold, provided there is sufficient gain available from the GS, lasing will occur at the GS and this will “clamp” its occupation at the level of population inversion required to overcome the laser cavity’s losses; any additional carriers injected into the GS beyond threshold will recombine quickly by stimulated emission and an increased light output. Under these conditions the X1 population should also be clamped and dual-state lasing will not occur. However a bottleneck will allow an increased X1 population, and rate-equation models based on this picture have successfully simulated the switch-on behavior of dual-state lasing. Such a simple model is appealing but the bottleneck argument assumes that the time-averaged GS and X1 populations can be described globally across the laser device and across the longitudinal and transverse optical modes of the laser cavity. Within these volumes there are typically millions of localized QDs, all randomly positioned and with varying emission wavelengths due to the inhomogeneous broadening of the QD ensemble. In addition, each laser cavity mode at the GS and X1 lasing wavelengths will have a different overlap with a different subset of the total QD ensemble and consequently a different optical loss. In considering the QD laser dynamics we argue here that it is important to consider the mechanisms that contribute to the collective behavior of the QD ensemble, both below and above the lasing threshold.

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Below the lasing threshold, the dominant mechanism for collective behavior within the QD ensemble will be carrier diffusion. Diffusion acts to equalize the carrier concentration across the device by the random movement of the electrons (and holes) which proceeds by thermal escape and subsequent re-capture into another dot. Thus more carriers escape from the heavily-occupied dots and the concentration tends towards an average level of occupation. Without carrier escape from the QDs, there is no mechanism for imbalances in occupation between neighboring dots to be corrected and the carrier distribution within the QDs becomes non-thermal, as is observed at low-temperatures [8, 9]. At higher temperatures, where thermal escape occurs and inter-dot diffusion proceeds, QDs are able to exchange particles and will do so in order to maximize the entropy of the system, thus defining the chemical potential of the system. In a semiconductor there are two chemical potentials, one each for the electrons and holes, which are referred to as quasi-Fermi levels. In an ideal diode the quasi-Fermi level separation at the junction is equal to the voltage applied to that junction by an external circuit, so the current-voltage (IV) characteristics of the diode junction in a QD laser can provide information on the collective behavior of the carriers in the QD ensemble. In this work, we compare the IV response of a quantum well (QW) and a QD laser, applying a small modulation signal in order to detect the onset of different laser modes and also the degree of clamping that is associated with them. This indicates that filamentation, governed by carrier diffusion amongst the QD ensemble, is responsible for dual state lasing, and this is supported by spectrally-resolved measurements of the mode profile obtained from the QD laser.

2. EQUIVALENT CIRCUIT MODEL FOR A QD LASER

We begin with an equivalent circuit model of the QD laser, shown in figure 1. Below the lasing threshold we assume that the active region of the QD laser behaves as an ideal diode ($Q_T$) in series with an ohmic resistance ($R_S$) which is due to the contacts, cladding layers and substrate of the laser, as shown in figure 1(a). The subscript “T” indicates the thermal nature of the quasi-Fermi levels which are important for the IV characteristic of the diode. Note that there is also an extra circuit element denoted by $Q_A$ which has been added to the circuit of figure 1(a). This represents the “rising-background” signal observed in modulation measurements [10], which we attribute to an “athermal” effect (hence the “A” subscript) due to the presence of the QDs; the need for this will become clear in the following section. When the laser threshold is reached, stimulated emission will clamp the occupation of the lasing state and this transforms figure 1(a) into figure 1(b) where $Q_T$ is replaced by a fixed voltage, equal to the diode junction voltage at the threshold.

Real QD laser: Clamped and unclamped regions co-exist in parallel

Figure 1: Proposed equivalent circuit models for the QD laser. (a) Sub-threshold; (b) Lasing; (c) Lasing and non-lasing filaments in parallel.
To form the equivalent circuit model for the real device requires careful consideration of the lasing properties of the QDs. The QD ensemble has a large inhomogeneous broadening and laser oscillation will occur when the gain of some portion of the dots balances the cavity losses. At that point, all QDs with emission energies that lie within the homogeneous linewidth will couple to this laser mode, *if they overlap well with the spatial extent of the mode*. This creates two interesting constraints on the QD laser: firstly, the homogeneous linewidth is known to be equal to or less than 10 meV at room-temperature whereas the full-width at half-maximum of the ensemble photoluminescence emission from the GS is always seen to be >20 meV and only a subset of the dots can be involved in the laser oscillation, secondly, the spatial extent of the optical modes of the laser cavity will not overlap completely with the electrically-pumped area of the device, as illustrated in Figure 2. Optimizing the spatial overlap of the optical modes with the electrical injection is a vital part of laser structure design and is hampered by current spreading [11]. The result of these two constraints is that some dots will be involved in the lasing and some will not, so the equivalent circuit for a real QD laser should include at least two current paths, one clamped and one diode-like (sub-threshold) as shown in figure 1(c). Below threshold it will behave identically to the circuit of figure 1 (a) as a normal QD diode; above the lasing threshold \( Q_{T2} \) will become clamped. Any increase in the bias current would require an increased voltage drop across \( Q_{T1} \) compared to \( Q_{T2} \), causing an imbalance in the circuit loop. As a result a current will flow around the loop from \( Q_{T1} \) to \( Q_{T2} \). This redirection of the electrical current, so that it passes through the region of the laser overlapping with the optical mode, will be referred to hereafter as filamentation; the term “filament” will then signify an identifiable current-path through the laser device. Filamentation will reduce but not eliminate the diode-like IV characteristics from the IV curve of the laser and will affect the series resistance and “athermal” contributions to the IV curve.

Figure 2: An optical schematic of the laser device: (a) Left, below-threshold the electrical-injection may be considered to be even across the device; (b) Center, above-threshold with a single transverse mode the overlap of the smaller optical mode with the electrically-injected area is incomplete; (c) Right, multi-mode lasing allows several optical modes to gain optimal overlap with the electrically-injected region.

It is expected that the QD-like athermal contribution from \( Q_{A2} \) will be increased because it is no longer in parallel with \( Q_{A1} \), as it was below threshold. Figure 2 outlines what happens optically within the laser for the different cases just described. Below the lasing threshold, light is spontaneously emitted by all dots, case (a). At threshold the overlap
between the large region of electrical injection and the optical mode is incomplete. Current will migrate to the lasing mode because the diode potential is clamped, while the contribution of the other IV characteristics would be increased by the concentration of current into the smaller filament. However if the laser exhibits multiple modes, particularly multiple transverse modes of the cavity (with their large differences in overlap with the active region), then figure 2(c) is the appropriate picture. In this case the area of the combined lasing filaments is increased compared to the single-mode case, so when a transverse mode appears in the laser emission, the diode-like characteristic is clamped more strongly while the other IV characteristics will decrease as the current is now directed into a larger area once more.

This model could apply equally well to a quantum well (QW) laser (with the possible exception of the athermal component of the IV curve to be introduced later). The main difference is in the lateral diffusion length, which is much greater for the QW laser than the QD laser [12]. Lateral diffusion of carriers would be expected to be the dominant mechanism of current re-direction between filaments which is indicated by the resistor bridging the two filaments in figure 1(c). The diode’s cladding regions and contacts should have a comparatively low resistance, making it difficult to for the current paths to form into filaments until they reach the intrinsic active region of the laser. This indicates that the series resistance may exhibit a more complex behavior than the athermal effects.

### 3. COMPARING QD AND QW LASER CHARACTERISTICS

#### 3.1. Devices under test

To illustrate the differences between a homogeneous QW laser and the inhomogeneously-broadened QD laser, a commercial MQW laser operating at ~980 nm was characterized for its optical and electronic properties alongside a QD laser. The QD laser incorporated three GaAs-capped QD bilayers [13], emitting at 1340 nm at room temperature in a ridge waveguide structure, 15 μm wide by 5 mm in length (for a more detailed description of the QD laser structure, see reference [10]).

#### 3.2. Characterization techniques

The characterization was performed in DC mode, the current (I) was supplied to the device-under-test (DUT) by a Thorlabs LDC205 current source; the luminescence (L) was monitored by an integrating sphere and InGaAs photodiode, whose photocurrent was measured by a Keithley 2520 laser test system that also monitored the DC voltage drop (V) across the DUT. Measurements of L and V across a range of I were made and are referred to hereafter as either LI, IV or LIV curves. Optical spectra were gathered as a function of current by directing the luminescence into a Spex 500M monochromator via a multimode optical fiber from the integrating sphere, where it was detected by a liquid nitrogen cooled Ge photodiode using standard lock-in techniques.

In addition to the standard LIV characterization, −I^2d^2V/dI^2 measurements were performed using modulation spectroscopy. To measure dV/dI, a modulation signal at ~15 kHz was supplied to the modulation input of the current source and the AC voltage response from the DUT was measured using a lock-in amplifier. However, measuring d^2V/dI^2 was complicated by the need to avoid harmonic interference; this was achieved by adding a second modulation signal at ~16 kHz to the modulation signal and measuring the voltage response, using a lock-in amplifier, of the DUT at the difference frequency of ~1 kHz. The prefactors of I and −I^2 are then added to the data to ease interpretation of the results, which can be illustrated by considering the IV curve of an ideal diode with a series resistance (1) and comparing with IdV/dI (2) and −I^2d^2V/dI^2 (3) measurements.

\[
V = \frac{\eta k_B T}{q} \ln \left( \frac{I}{I_S} + 1 \right) + IR_S 
\]

\[
I \frac{dV}{dI} \approx \frac{\eta k_B T}{q} + IR_S
\]
Here $q$ is the charge on an electron, $k_B$ is Boltzmann’s constant, $T$ is the temperature in Kelvin and $R_S$ is the series resistance. $I_S$ is the saturation current and is equal to the theoretical reverse-bias leakage of an ideal diode, while in forward-bias it determines the switch-on voltage. $I_S$ is small compared with $I$ at the current-levels being investigated here and can be neglected. $\eta$ is known as the “ideality factor” which, for conventional bulk or quantum well devices, is unity for “ideal” Shockley recombination (e.g. radiative), ~2 for Hall recombination via traps (or an intermediate value if both processes operate) or greater than 2 in a diode with a degenerate active region [14]. Therefore, when the $I$\textsuperscript{dV/dI} measurement is performed on an ideal diode the ideality factor and series resistance can be extracted, which shows that this technique is useful for analyzing the details of the device behavior [15]. The $-I$\textsuperscript{2d^2V/dI^2} measurement is more sensitive to the diode junction’s behavior and is independent of the series resistance $R_S$; this measurement should give a constant signal equal to the ideality factor of the diode below the lasing threshold.

3.3. Characterization results

Figure 3 compares the results of the characterization of the QW and QD bilayer lasers. The LI curve of the QW laser indicates weak spontaneous emission below the lasing threshold and a linearly increasing laser emission above threshold. Above the point marked 6 (or region vii) there is a slight roll-over, normally attributed to device heating. The corresponding IV curve is diode-like below the lasing threshold becoming linear (Ohmic) above due to clamping of the quasi-Fermi level separation when laser oscillation occurs.
The $-I^2d^2V/dI^2$ measurement shows a constant signal up to the laser threshold, as expected for an ideal diode junction from equation (3), followed by a sharp peak at the lasing threshold. The signal then drops to zero, consistent with the clamping of the quasi-Fermi levels causing the diode-like characteristic of the IV curve to be suppressed under lasing conditions. At higher currents two small peaks (points 2 and 3) and a larger disturbance (point 4) are seen. The latter feature correlates with a shift in the spectrum of the laser (figure 3(e)) which we can attribute to a change in the mode pattern of the emission [10]. It can also be seen that in region (vii) the curve becomes non-zero again, suggesting that the quasi-Fermi levels are no longer completely clamped. This correlates with the roll-over of the LI curve and suggests that device heating is not responsible. Thus the combination of electrical and optical measurements provides us with a good explanation of the behavior of the QW laser.

Figure 3: (top) An $-I^2d^2V/dI^2$ measurement taken at 17 °C. This has been arranged as a horizontal slice (indicated by the arrows, with the original y-axis becoming intensity) in an image (bottom) to illustrate the temperature dependence of the dual-state lasing for the QD lasers.
Turning to the QD laser we see similar features but a major difference; the onset of dual-state lasing at ~290 mA. In addition, the IV curve shows no obvious features at either the laser threshold or the dual-state lasing threshold and does not become Ohmic. The $-\Delta V/dI^2$ measurement suggests further differences between the QW and QD laser. The latter shows a clear clear intercept voltage at low current, indicative of an ideal diode characteristic but the signal constantly rises, almost linearly. This rising background signal is the athermal characteristic that was included in the equivalent circuit model and is found to be present in all the QD diodes we have investigated (our own samples and those grown by other groups). There are many peaks of varying sizes at the lasing and dual-state thresholds but also at intermediate points. Each peak is followed by a drop in the $-\Delta V/dI^2$ signal (indicating that at least partial quasi-Fermi level clamping is occurring) but none of the drops is large enough reduce the $-\Delta V/dI^2$ signal to zero (as it did for the QW laser).

We note that the dual-state lasing threshold has the same general form as the initial lasing threshold and even the intermediate peaks. We see that these latter peaks are associated with small kinks in the LI curve and we attribute these to filamentation events due to small transverse mode changes.

### 3.4. Temperature dependence of the dual-state lasing

We have investigated the temperature dependence of the dual state lasing using the methods described above in order to refine our model. By taking a single $-\Delta V/dI^2$ measurement as a horizontal slice, then repeating the measurement for a range of different temperatures and stacking them into a 2-D composite we obtain figure 4. Peaks due to filamentation events can clearly be seen, and the figures show their evolution over the temperature range 10 – 45°C for a range of drive currents.

We observe that the onset of GS lasing rises to higher currents with increasing temperature until eventually reaching a point where the X1 lasing threshold catches up with it; to higher temperatures only X1 lasing is observed. These data suggest that the various modes/filaments present in the laser are competing with each other as conditions are varied. For example, when GS lasing is present it competes with the dual-state/X1 lasing threshold by delaying its onset until higher currents. This behavior is well-documented in previous reports of dual-state lasing [5].

When the X1 mode takes over from the GS lasing the filamentation peak for the X1 lasing threshold becomes much larger (also seen when one of the smaller GS peaks “merges” with the dual-state lasing threshold). This also supports the argument that these modes are competing spatially. These data also illustrate the utility of the $-\Delta V/dI^2$ measurement in analyzing the modal dynamics of QD lasers, by charting out the temperature dependence as shown in figure 4, regions of interest can readily be identified for study by other techniques, for example, profiling the laser emission.

### 4. ANALYSIS OF THE FILAMENTATION IN THE QD LASER

Having established that the QD laser exhibits dual-state lasing and with the aid of the equivalent circuit model, the filamentation events in the laser can be analyzed with some degree of quantitative rigor. To do this, linear fits were made, where possible, to the $-\Delta V/dI^2$ measurement between the filamentation peaks. An example of this fitting scheme is shown in Figure 5(a) below. A linear fit to the $-\Delta V/dI^2$ measurement below the lasing threshold was used to identify the offset in the signal due to the ideal diode-like characteristic $\eta k_BT/q$. Additional linear fits between the filamentation peaks were then used to determine the gradients of the QD-like athermal characteristic as well as evaluating the changes in the ideal diode characteristic at each filamentation event, $\Delta \eta k_BT/q$ and $\Delta \eta k_BT/q$. The changes in the ideal diode characteristic are plotted in Figure 5(b), where the squares indicate the ideal-diode characteristic and how it rises slightly with increases in temperature. The circles indicate the diode-like characteristic that is present after the GS threshold, while the triangles indicate how much remains after the X1/dual-state lasing threshold.

Figure 5(b) shows that only partial clamping of the diode-like characteristic is observed when only the GS or X1 states are lasing, except at the highest temperatures (where carrier diffusion due to escape should be the strongest). However, a high degree of clamping was observed for dual-state lasing on both GS and X1, with complete clamping near 30 °C. The observation that neither the GS nor X1 are able to fully-clamp the device at lower temperatures indicates that neither of the optical modes that achieve lasing have a complete overlap with the electrically-injected region of the laser. The degree of clamping by the X1 mode is however, much larger than for the GS mode; this also indicates that the two...
modes are not orthogonal but overlap significantly, as supported by their competition in the temperature dependence data. This partial clamping behavior is fully consistent with the expectations of the equivalent circuit model proposed above suggesting that spatial competition by carrier diffusion is a sufficient model to explain the switch-on behavior of the dual-state lasing phenomenon.

Figure 4: (a) An $-I^2d^2V/dI^2$ measurement (solid line) with an example of the fitting process overlaid; the dotted-lines indicate the linear fits made between the filamentation peaks at 100 and 245 mA, the intercept $\eta k_B T / q$ (the ideal diode-like characteristic) is highlighted, along with the changes to it at the lasing thresholds, $\Delta \eta_1 k_B T / q$ and $\Delta \eta_2 k_B T / q$. (b) The results showing the diode-like characteristic below-threshold, above the GS threshold and above the X1/dual-state threshold. (c) The gradients of the linear fits made between the filamentation events.

The gradient data shows that when the GS mode reaches the laser threshold, the gradient increases sharply but then decreases after each subsequent filamentation event, such as the onset of additional transverse modes and dual-state...
lasing, eventually returning to its sub-threshold level. As the gradient has units of resistance, this suggests that the initial filamentation event is concentrating the injected current into a smaller area of the device (i.e., the initial filament is smaller than the area of electrical-injection). This is supported by the observation that, at the higher temperatures where only the X1 mode achieves lasing, the gradient is also higher than the sub-threshold value. These data again indicate that the GS and X1 modes occupy an area smaller than that for electrical-injection into the laser. This concentration effect into a small area when the first mode lases, and which is subsequently relaxed as more modes of the device switch on and the electrically-injected area is utilized more completely, is fully consistent with the equivalent circuit model presented earlier.

From these combined data sets we conclude that different areas of this QD laser are involved in lasing on different states and that the “bottleneck” is in fact due to carrier diffusion not being strong enough to couple the electrical injection into the lasing mode(s). If this is the case, transverse modes are likely to be responsible because they provide a large difference in spatial overlap, so we proceeded to investigate the mode-pattern of the emission in more detail.

5. SPECTRALLY-RESOLVED MODE PROFILING

The mode profile of the QD laser emission at the dual-state threshold has been investigated briefly in the literature [10, 16] and it was noted that there is a switch, or destabilization, in the emission pattern when the phenomenon occurs. Here the effect was investigated with a scanning pinhole spatial filter, coupled into an InGaAs array spectrometer, which could resolve the laser emission’s far-field both spatially and spectrally. Apochromatic NIR optics were used to avoid distortions due to chromatic aberration. This is necessary because the GS and X1 are separated in wavelength by 100 nm and the spatial filter was required in order to obtain a consistent profile of the lasing emission from both states. The DUT for this measurement was a 3 mm by 5 μm wide device with five QD bilayers. The results of measurements taken at 500 and 800 mA are shown in figures 6 and 7, respectively.

Figure 6: The spectrally-resolved mode profile of the QD bilayer laser when operated at 500 mA bias current. The GS is the dominant source of emission and shows a strong lobe in the centre-right of the image and a weaker side lobe to its left. The X1 emission is centered on the minimum between the two GS lobes and is clearly resolved from the GS mode profile.

The GS and X1 modes are clearly resolved; the X1 mode has a larger spatial extent and also overlaps with the GS mode, showing that they are not orthogonal. Interestingly, in figure 7 the weaker X1 mode appears where there is a minimum in the GS mode pattern, as might be expected if they are competing. This shows that in this laser the dual-state lasing occurs on different transverse modes, supporting the idea that dual-state lasing occurs not because of a bottleneck within
the QD electronic structure, but rather because the carrier diffusion between different QDs is unable to fully-equilibrate the ensemble.

Figure 7: The spectrally-resolved mode profile of the QD bilayer laser when operated at 800 mA bias current. The profile is similar except that the GS and X1 intensities are comparable, the GS emission is now a single lobe on the right; while the X1 mode is centered on the image, showing its larger spatial extent and overlapping with the GS mode.

6. CONCLUSIONS

We propose that when considering a QD laser or other device, collective behavior should be evaluated by considering the coupling mechanisms that give rise to it. In the case of room-temperature QD lasers, inter-dot carrier diffusion by escape and re-capture is the dominant mechanism for creating a quasi-Fermi level across the device. The phenomenon of dual-state lasing was introduced and its interpretation as a bottleneck, invoked to de-couple the GS and X1 states, discussed. An equivalent circuit model was formulated to represent the QD laser and the importance of co-existing lasing and non-lasing filaments in the device considered. This model is valid for both QD and QW (where diffusion is known to be stronger) lasers and so they were compared and the differences illuminated.

The filamentation events in the QD laser that occur for dual-state lasing were investigated in detail: the temperature-dependence was illustrated and then the changes at each threshold were analyzed. It was found that the GS laser mode appears to partially clamp the diode-like characteristic of the IV curve and suggests that current is being concentrated into a smaller area of the device. When the X1 lasing occurred simultaneously, the clamping was more complete but the concentration effect was reduced. This was interpreted in terms of different spatial regions being responsible, in this laser, for the GS and X1 modes. Spectrally-resolved mode profiling of another laser confirms that the filamentation at the dual-state threshold corresponds to the X1 lasing occurring on a different transverse mode of the laser, consistent with the IV curve measurements and equivalent circuit model.

These results suggest that the equivalent circuit model, which suggests that dual-state lasing is due to a “bottleneck” 

between different dots, provides a sufficient model to explain the onset of dual-state lasing in QD lasers. The idea of a phonon-bottleneck within the dots relies on the ability of carrier diffusion to maintain equilibrium across the entire device because it assumes a global population level. It is therefore not invalid but this work indicates that the spatial inhomogeneity of the QD ensemble carries at least equal importance in considerations of QD laser dynamics.
ACKNOWLEDGEMENTS

The authors wish to thank Daniel Farrell for many useful discussions and the Quantum Photovoltaics at Imperial College London for assistance with equipment. This work was funded by the Engineering and Physical Sciences Research Council, U.K.

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