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Experimental Evidence of the Impact of Nitrogen on Carrier Capture and Escape Times in InGaAsN/GaAs Single Quantum Well

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Abstract: We report our experimental results and theoretical analysis on carrier escape time in $In_{0.4}Ga_{0.6}As_{1-y}N_y/GaAs$ (y = 0; 0.005) ridge waveguide single-quantum-well (QW) lasers with N-contents of 0% and 0.5%. The experiments were carried out by using novel time-resolved two-color pump–probe transmission measurements. Our results show a significant decrease of carrier escape time with nitrogen incorporation in the InGaAsN QW, which agrees well with the values obtained from the theoretical calculation based on thermally activated hole leakage. The measurement results provide experimental supports for hole leakage theory.

Index Terms: Quantum-well (QW) lasers, carrier capture and escape process, InGaAsN, 1.3- μ m lasers.

1. Introduction

Dilute-nitride-based diode lasers have significant applications for uncooled telecom applications [1]–[9]. Significant progress has been realized resulting in low-threshold diode lasers operating at 1300–1500 nm for dilute-nitride-based active regions [1]–[14], resulting in low-temperature-sensitive lasers operating at this spectral regime. Several studies have been attempted to understand the dominant factors limiting the temperature sensitivity [10]–[14] and optical gain and threshold characteristics [15]–[23] of dilute-nitride lasers.

Recently, high-performance InGaAsN/GaAs quantum-well (QW) lasers have been demonstrated with high-speed modulation bandwidth [24], [25]. However, these benchmarks are still distanced from the high-frequency bandwidth limit predicted by theories [26], [27]. The carriers capture (τ_{cap}) and escape times (τ_{esc}) are very important parameters that determine the static and dynamic performances of QW lasers [28]–[30]. In dynamic operating conditions, the capture and escape

times affect both the resonant frequency and the damping rate of the modulation response [28]–[30], and thus determine the modulation bandwidth of the lasers.

It has been pointed out that the bandwidth of current dilute-nitride lasers are mainly limited by thermal effects that reduce the effective differential gain and lead to a rapid increase of the threshold current and reduction of external differential quantum efficiency [31], [32]. In addition, supportive theoretical calculations of carrier thermionic escape process have suggested the existence of an increased hole leakage, as a contributing factor to the device temperature sensitivity [33]. However, no direct experimental evidence on the hole leakage effect in InGaAsN QW laser has been provided so far. These calculations have relied on the assumption of a smaller valence-band offset in the InGaAsN QW lasers [33], which also contains significant uncertainty from experiments. Motivated by this necessity, in this paper, we investigate the carrier capture and escape processes (τ_{cap} and τ_{esc}) of InGaAsN/GaAs QW lasers under differential transmission measurements and time-resolved photoluminescence (TRPL) measurement. An identical laser structure with identical In-content InGaAs QW without any nitrogen incorporation is also measured as control sample.

From the experimental results, τ_{esc} with bias current for the InGaAsN QW laser show a different trend, and τ_{esc} for InGaAsN QW lasers are one order of magnitude smaller in comparison with those measured of InGaAs QW laser at higher current. The dependence of τ_{esc} on carrier density in dilute-nitride QW lasers for low-carrier-density operation is only explained if tunneling through the triangular-biased QW is considered. However, the behavior of τ_{esc} on carrier density in InGaAsN QW lasers at high carrier density can be explained well with the thermionic carrier escape model. The tunneling mechanism is most important at low carrier densities, as expected for a smaller band offset ratio. In contrast, the τ_{esc} characteristics of the InGaAs QW laser can be well explained by the thermionic carrier escape model for the wide range of carrier density investigated here.

2. Experimental Works

The carrier escape and capture processes were investigated on two identical $In_{0.4}Ga_{0.6}As$ and $In_{0.4}Ga_{0.6}As_{0.995}N_{0.005}$ single-QW ridge waveguide (RWG) lasers. Both of the lasers were grown on GaAs substrates by low-pressure (200 mbar) and low-temperature (530 °C) metal–organic chemical vapor deposition (MOCVD), with the details of the growths provided in reference [34]. The two device structures consist of a 6-nm single-QW active layer sandwiched by two tensile strain GaAs_{0.85}P_{0.15} barriers, a 300-nm GaAs separate confinement region (SCH), and AlGaAs claddings. The uncoated RWG InGaAsN (InGaAs) devices feature a stripe width of 3.5 μ m and a cavity length of 500 μ m, with emission wavelengths of $\lambda = 1275$ nm (1195 nm). The threshold current at room temperature were measured to be $I_{th} = 18$ mA (10 mA).

All previously reported two-color pump–probe transmission measurements required a high-power laser oscillator to generate a broadband continuum to serve as probe beam other than pump laser beam [35]–[37]. Aside from the experimental difficulty, such high power was prone to disturb the equilibrium of the measured system and make the data interpretation more challenging.

Here, instead, as shown in Fig. 1, we utilized residue light from a 800-nm mode-locked Tisapphire pulse laser as pump beam, and the probe pulse train was generated from an optical parametric oscillator (OPO), which was synchronously pumped by the mode-locked Ti-sapphire pulse laser, which, for the first time, allow direct measurement of carrier capture and escape processes in InGaAsN/GaAs QW structures. The Ti-sapphire oscillator emitted 100-fs pulses centered at a wavelength of 800 nm with 82-MHz repetition rate and average power of 900 mW. The average output power of the OPO was about 40 mW, with a pulsewidth of 150 fs. The wavelength of the OPO was tuned to the corresponding emission wavelengths of the laser devices under study, i.e., 1280 nm for InGaAsN QW lasers and 1200 nm for InGaAs QW lasers, respectively. Using an ND filter, the power of the pump beam was chosen to be around 3 mW and ten times larger than that of the probe beam. Relative delay between the pump and the probe was controlled through a steppingmotor-driven translation stage. Two microscope objectives were used to couple light in and out of the



Fig. 1. Experimental set up for two-color pump-probe transmission measurements. BC: beam combiner. A: current source.

waveguide of the RWG laser devices. Taking advantage of the different wavelengths of the pump and the probe, we eliminated the pump pulses by using a 1150-nm long-pass filter (LPF) before the final detection, and the change in probe transmission were detected as a function of pump–probe delay. Here, the lock-in technique was used, where only pump beam was chopped and the probe beam was measured at chopper frequency.

The carrier transport and carrier capture processes were investigated through the temporal evolutions of photoluminescence (PL), which employed a luminescence upconversion setup. The detail of the temporal evolution PL setup was described in our previous works [38].

3. Results and Discussion—Analysis

Fig. 2 shows the change of probe transmission, i.e., ΔT , induced by the pump as a function of pump-probe delay with different biasing conditions. An exponential decay following an instantaneous rise was observed for both lasers (see Fig. 2), and the carrier lifetimes were fitted following an exponential relation of $\Delta T = \Delta T_0 \times \exp(-t/\tau)$. With the pump beam exciting only the unconfined state outside of the QW and probing the ground state in the well, the rise time of ΔT reflects the process of carrier capture into the well. The decay time can be ascribed to contribution of both carrier escape from the well and carrier recombination. The latter, from our results of PL lifetime measurements at room temperature, is on the order of 300-400 ps for InGaAsN and > 2 ns for InGaAs lasers, respectively, as shown in the TRPL spectra comparison of In0.4Ga0.6As0.995 N0.005 QW and $In_{0.4}Ga_{0.6}As$ QW samples performed at T = 300 K (see Fig. 3) with carrier density of n \sim 2 \times 10¹⁸ cm⁻³. Note that the carrier recombination lifetimes in the QW include the contribution from both the radiative and nonradiative (Auger and monomolecular) recombination processes, and these total carrier lifetimes are relatively long in comparison with the decay rate of ΔT from the probe transmission measurements. The Auger process is a strong carrier-dependent process for high-carrier-density operation near threshold conditions; however, the total carrier lifetimes are found to be an order of magnitude larger in comparison with the decay rate ΔT . Thus, the decay of ΔT observed here mainly represents the carrier escape dynamics. Note that the samples were not biased in the TRPL measurements and the escape process was highly dependent on carrier density in the QW. In our TRPL measurements, the measured carrier lifetime is a combination of both the radiative and nonradiative recombination processes, as well as possible escape processes. In the pump-probe experiments, the device was biased with a perturbation introduced from the pump beam, which led to the probe revealing the differential carrier lifetimes. Previous data on the strong dependence of the external differential guantum efficiency with temperature is indicative of carrier leakage [10], [13], [39], since Auger recombination should only impact the temperature dependence of the threshold current [16].

A more significant decrease of decay time with increased bias can be observed for the dilutenitride laser, compared with the InGaAs laser. At a bias just below threshold, τ_{esc} is found to be one



Fig. 2. Relative changes in the probe transmission induced by the pump as a function of the pumpprobe delay, for both InGaAs and InGaAsN RWG lasers. The biasing is varied below threshold. The traces are recorded at the wavelengths indicated in the figure.



Fig. 3. TRPL spectra at T = 300 K for $In_{0.4}Ga_{0.6}As_{0.995}N_{0.005}$ QW and $In_{0.4}Ga_{0.6}As$ QW samples. The solid black traces with arrows are time integrated PL spectra.

order of magnitude lower in InGaAsN laser as compared with that measured from the InGaAs laser sample. The smaller τ_{esc} will lead to reduced current injection efficiency, as well as a reduction of the modulation bandwidth of the lasers [40]–[42].



Fig. 4. Experimental (points) and calculated (thermionic processes: solid lines; tunneling: dotted lines) carrier escape times as function of carrier density for InGaAs and InGaAsN lasers.

We performed a thermionic escape time calculation to understand the underlying reason for the decreased τ_{esc} in dilute-nitride laser. Obtaining the relationship between the bias I and carrier density N, the decay time as function of carrier density is plotted in Fig. 4. The relationship between I and N is extracted by measuring the gain spectra and fitting it with an analytical gain model [43].

Treating the carrier escape process via the classical thermionic emission model and considering the state filling effect, the functional dependence of $\tau_{esc}(N)$ can be calculated using the following analytical equations [33], [44], [45]:

$$\tau_{\text{esc_e}}(N) = \omega \frac{L_{qw}}{m_{e_b}} \left(\frac{\pi \hbar^2 m_{e_w}}{m_{e_b} k_B T} \right)^{\frac{1}{2}} \frac{\exp\left(\frac{qV_e}{k_B T}\right)}{1 + \exp\left(\frac{E_{fc}(N) - E_{e_1}}{k_B T}\right)}$$
(1)

$$\tau_{\text{esc_hh}}(N) = \omega \frac{L_{qw}}{m_{\text{hh_b}}} \left(\frac{\pi \hbar^2 m_{\text{hh_w}}}{m_{\text{hh_b}} k_B T} \right)^{\frac{1}{2}} \frac{\exp\left(\frac{qV_{\text{hh}}}{k_B T}\right)}{1 + \exp\left(\frac{E_{lv}(N) - E_{\text{hh_1}}}{k_B T}\right)}$$
(2)

$$\frac{1}{\tau_{\text{esc}}(N)} = \frac{1}{\tau_{\text{esc}_e}(N)} + \frac{1}{\tau_{\text{esc}_hh}(N)}$$
(3)

where ω is a constant related to the escape rate from inside the well to unconfined state in the barrier, in units of $m^{-1} \cdot kg \cdot eV^{-1/2}$, and we assume that ω remains identical for both InGaAs and InGaAsN, L_{qw} is the width of the QW, $m_{e_{-W}}$ ($m_{hh_{-W}}$) is the effective electron (heavy hole) mass in the well, and $m_{e_{-b}}$ ($m_{hh_{-b}}$) is the electron (hole) mass of the barrier. qV_e and qV_{hh} are the energy difference between the first confined electron (heavy hole) state and the edge of barrier conduction and valence bands, respectively. $E_{fc}(N)$ [$E_{fv}(N)$] corresponds to the electron (hole) quasi-Fermi level for confined carriers, and $E_{e_{-1}}(E_{hh_{-1}})$ is the energy level of the first confined level.

Note that, for the case with more than one confined energy level, the escape time is replaced by a summation over all energy levels involved. For the carrier densities of our interest ($< 3 \times 10^{18} \text{ cm}^{-3}$), the quasi-Fermi level is far below the second confined level for both electron and holes; thus, the corresponding exponential term in the denominator is much less than 1 and is negligible. Table 1 summarizes all the parameters used in our paper.

The results of the calculation are plotted in Fig. 4, together with the relaxation times determined experimentally. The solid lines are the results for free-carrier thermionic escape, where the difference between InGaAs and InGaAsN lasers in the calculation are only the electron effective mass and band offset ratio. For a value of $\omega = 2.1 \times 10^{-21} \text{ m}^{-1} \cdot \text{kg} \cdot \text{eV}^{-1/2}$ chosen, the escape time of one order less is obtained for InGaAsN showing good agreement from other theoretical calculations

	$In_{0.4}Ga_{0.6}As_{0.995}N_{0.005}/GaAs$	In _{0.4} Ga _{0.6} As/GaAs
ΔE_{c}	431 meV	251 meV
$\Delta E_{ m v}$	95 meV	177 meV
qVe	360 meV	177 meV
qV_{hh}	85.6 meV	163.5 meV
m _{e w}	0.11 m ₀	0.047 m ₀
m _{e b}	$0.067 \ m_0$	0.067 m ₀
m _{hh w}	0.465 m ₀	0.465 m ₀
m _{hh b}	0.51 m ₀	0.51 m ₀
ω (free carrier)	$2.1 \times 10^{-21} \mathrm{m}^{-1} \mathrm{kg} \mathrm{eV}^{-1/2}$	$2.1 \times 10^{-21} \mathrm{m}^{-1} \mathrm{k} \mathrm{g} \mathrm{eV}^{-1/2}$
ω (exciton)	$5 \times 10^{-21} \mathrm{m}^{-1} \mathrm{kg} \mathrm{eV}^{-1/2}$	$5 \times 10^{-21} \mathrm{m}^{-1} \mathrm{kg} \mathrm{eV}^{-1/2}$

 TABLE 1

 Band parameters used in the simulations of the tunneling and thermionic carrier escape times for both InGaAs QW and InGaAsN QW [42], [43].

on the same laser structures reported previously [28]. Notice that, while the calculation gives similar values at higher carrier density, the function trend is off at low carrier density, especially for InGaAsN.

The discrepancy between the measurements of τ_{esc} for InGaAsN laser and the results from the thermionic simulation indicates that there are other mechanisms that dominate the carrier escape at low carrier density [40]–[42]. One such possible carrier escape mechanism at low carrier density is the tunneling phenomena. From the carrier tunneling rate expressions in (4) and (5) [42], one finds that the carrier escape mediated by the tunneling process has a stronger dependence on carrier density, which shows that a faster decay rate with increased carrier density (index of 3/2 in exponential term) would be expected as compared with the thermionic model with exponential index of 1 for electrons and holes, as follows:

$$\tau_{\text{tun}_e}(N) \sim \exp\left(\frac{4}{3} \frac{\sqrt{2m_e^*}}{e\hbar F(N)} (V_e - (E_{fc}(N) - E_{e_1}))^{\frac{3}{2}}\right)$$
(4)

$$\tau_{\text{tun_hh}}(N) \sim \exp\left(\frac{4}{3} \frac{\sqrt{2m_{\text{hh}}^*}}{e\hbar F(N)} (V_e - (E_{\text{fv}}(N) - E_{\text{hh}_1}))^{\frac{3}{2}}\right).$$
(5)

In the above expressions, F(N) is the built-in field resulting from the biasing, which contributes to the decrease in escape time with carrier density. Assuming that the impedance of the laser diode does not change with bias, the built-in field is proportional to the biasing current.

The dotted line in Fig. 4 is the simulation result considering only tunneling in the InGaAsN QW laser. All the parameters used in the fitting are the same as those in Table 1. Note that the thermionic carrier escape rates [42] primarily depends on the conduction- and valence-band offsets for respective QW structures, as well as the corresponding effective masses for both carriers in respective QWs. In our studies, the band offsets and corresponding effective masses for InGaAs QW and InGaAsN QW were taken from references [42] and [43]. In our paper, the structures consist of InGaAs QW and InGaAsN QW with direct GaAs barriers followed by tensile GaAsP barriers for strain compensation purposes. Thus, the direct heterostructures studied here consist of InGaAsN QW is remarkable and explains the steep decay of τ_{esc} clearly showing the dominance of carrier escape out of the well through tunneling at low-carrier-density regime. As carrier density increases in InGaAsN QW, both thermionic escape and tunneling become important. However, at the very high

carrier density in the InGaAsN QW, the carrier escape in InGaAsN QW appears to be sufficiently described by the thermionic carrier escape model.

The analysis and experimental results of this paper shows that the thermionic carrier escape model taking into consideration the hole leakage provides good agreement with the experimental data for InGaAsN QW lasers. These results agree well with the previously reported studies on the possibility of hole carrier leakage in InGaAsN QWs [33], [42], [46]–[48]. The use of GaAsP direct barriers with larger band offset to surround the InGaAsN QW had resulted in a reduction in the threshold current density and an increase in the differential quantum efficiency for strain-compensated dilute-nitride lasers [13], [42]. In addition, recent theoretical works by Healy and O'Reilly [16] had also shown that the implementation of GaAsP barriers leads to improved hole confinement in the InGaAsN QW. However, it is important to note that other possible leakage mechanisms in InGaAsN QW that can lead to thermionic carrier escape processes. Recent findings by Chamings *et al.* have also suggested the possibility of carrier leakage process via defect states recombination in barrier regions in dilute-nitride QW structure [49].

Note that the results for the InGaAs QW lasers indicate that the carrier escape processes appear to be dominated by the thermionic carrier escape, as supported by the good agreement between the theoretical model and experimental results for InGaAs QW (see Fig. 4). Furthermore, note that the thermionic carrier escape rate appears to be an order of magnitude lower than that of InGaAsN QW.

In the analysis of the current injection efficiency and modulation bandwidth in QW lasers, both the carrier escape time (τ_{esc}) and carrier capture time (τ_{cap}) are of importance. Generally, it is not just the value of carrier escape time but the ratio of capture and escape time constant ($R = \tau_{cap}/\tau_{esc}$) that determines the modulation response and current injection efficiency in QW laser. Note that the τ_{cap} is the sum of quantum capture time $\tau_{cap_{qw}}$ and the transport time from the edge of barrier to the QW, i.e., $\tau_{\rm tr}$. The rise time in our pump-probe data indicates that the quantum capture time $\tau_{{\rm cap}_{qw}}$ is less than 1 ps for both InGaAsN and InGaAs QWs. In our studies, τ_{tr} was calculated by using the approach in reference [32], and τ_{tr} was found as constant \sim 5.3 ps in both InGaAsN and InGaAs lasers. In addition, we also determined the transport time as \sim 10 ps from TRPL rise time. Thus, we conclude that transport time is the dominant contribution to τ_{cap} in both lasers and that the introduction of nitrogen does not affect τ_{cap} significantly. This finding is consistent with general knowledge on the physics of QW lasers. The measurements of the distinct differences in thermionic carrier escape processes in InGaAs/GaAs QW and InGaAsN/GaAs QW lasers point out to the importance of taking into account the carrier escape processes in QW LEDs and lasers, and the finding and method described here are useful for providing improved understanding of the characteristics of electrically injected other QW LEDs and lasers operating at high current density including for providing insight into the efficiency-droop phenomenon in InGaN-based QW devices [50]-[56].

4. Summary

We have independently measured carrier escape times τ_{esc} for both InGaAsN and InGaAs QWs lasers. From our experiments, we found that τ_{esc} in InGaAsN QW laser is almost an order of magnitude lower in comparison with that measured in InGaAs QW laser. We show that the dependence of τ_{esc} for InGaAsN QW laser in the low carrier density regime can only be explained by taking into account the carrier escape process by tunneling phenomena. The tunneling carrier escape mechanism is most important at low carrier densities in InGaAsN QW due to the smaller band offset ratio, particularly for the valence-band offset. Thermionic carrier escape process is dominant for InGaAsN QW laser operating at high-carrier-density regime. In contrast, the carrier escape process in InGaAs QW laser can be well explained only with the thermionic carrier escape model for both low and high carrier density regimes. Together with TRPL measurements, the ratio of τ_{cap}/τ_{esc} of InGaAsN laser device was found to have strong carrier density dependent, while minimal-carrier-density-dependent τ_{cap}/τ_{esc} was observed for InGaAs QW lasers. The finding shows the importance of taking into consideration the effect of carrier escape processes on current injection efficiency in QW lasers or LEDs operating at high current density, which clearly indicates that the current injection efficiency needs to be accounted for in the analysis of internal quantum

efficiency and laser characteristics in QW devices [42]. In addition, these results provide improved understanding on the device physics of current injection efficiency in electrically injected QW devices, which is an imperative parameter for addressing solutions to achieve low-threshold lasers operating at high temperature and suppress efficiency-droop phenomenon in III–V or III-Nitride QW LEDs.

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