The Effect of Doping in Split-Well Direct-Phonon THz Quantum-Cascade Laser Structures

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Abstract: We have studied the effect of doping on the temperature performance of a split-well (SW) direct-phonon (DP) terahertz (THz) quantum-cascade laser (QCL) scheme supporting a clean three-level system. Achieving a system that is as close as possible to a clean n-level system proved to be the strategy that led to the best temperature performance in THz-QCLs. We expected to obtain a similar improvement to that observed in resonant-phonon (RP) schemes after increasing the carrier concentration from $3 \times 10^{10}$ cm$^{-2}$ to $6 \times 10^{10}$ cm$^{-2}$. Our goal was to improve the temperature performance by increasing the doping, ideally the results should have improved. To our surprise, in the devices we checked, the results show the contrary. Although an increase in doping had previously shown a positive effect in RP schemes, our results indicated that this does not happen with SW–DP devices. However, we observed a significant increase in gain broadening and a reduction in the dephasing time as the doping and temperature increased. We attribute these effects to enhanced ionized-impurity scattering (IIS). The observation and study of effects related to dephasing included in our experimental work have previously only been possible via simulation.

Keywords: THz-QCLs; doping effect; LO-phonon scattering

1. Introduction
Terahertz (THz) quantum-cascade laser (QCL) are noteworthy because of their range of potential applications, which includes nondestructive detection of materials, imaging, and gas spectroscopy [1–4]. These unique lasers are used in a variety of fields, including astrophysics [5], medicine [6,7], security [8], and chemistry [9,10]. Recently THz QCLs proved to be beneficial for wireless communication and imaging technologies [11]. Although THz QCLs were demonstrated in 2002 [12], their use in many of these fields has been restricted by the lack of portability of devices. In particular, there has been relatively little progress in increasing the maximum operating temperature ($T_{\text{max}}$) of the devices. This means special machinery is needed in order to reach cryogenic temperatures. Until recently, the $T_{\text{max}}$ reported for a GaAs/AlGaAs THz QCLs was 210 K [13]. In 2020, a $T_{\text{max}}$ of 250 K was achieved and demonstrated [14], enabling the launch of the first high-power portable THz QCL. This device, although portable, still required thermoelectric cooling. Further research and improvement in temperature performance has been reported in other devices, such as a down converted THz frequency comb, that can be applied to room temperature THz spectroscopy [15]. Our goal is to address those mechanisms that continue to limit device performance, thereby enabling room-temperature operation in the near future.

Some of the mechanisms limiting the temperature performance of THz QCLs have already been addressed in previous papers and their effect minimized. It is known that the main mechanism limiting the performance of vertical-transition THz QCLs is nonradiative thermally activated longitudinal-optical (LO)-phonon scattering from the upper laser level (ULL) to the lower laser level (LLL) [16]. This is significantly reduced in diagonal transition structures [17,18]. Further investigation has shown that thermally activated carrier leakage into the continuum [17] was very common when using barriers containing just 15% Al.
Leakage into excited bound states [19] was observed even with higher barriers (increased Al content). Carefully engineered designs that combined higher barriers with thinner wells were able to suppress these leakage mechanisms [19]. Unlike earlier devices, these devices showed clear negative differential resistance (NDR) behavior in their current–voltage (I–V) curves even at room temperature. The NDR signature meant that no leakage channels were activated at room temperature, in this way achieving what appeared to be a clean n-level system. Following this work, the strategy has been to achieve a system that is as close as possible to a clean n-level system. This was the strategy adopted in [14], where a $T_{\text{max}}$ of 250 K was reported, and this is the strategy we also adopted in our experimental work.

However, despite the NDR observed in [19], the temperature performance failed to improve further. This was attributed to the increased lifetime of the LLL and an increase in interface roughness (IFR) scattering [20–24]. More information about scattering mechanisms and transport characteristics of quantum lasers can be found in [25,26], as well as lifetimes of the different laser levels in [27]. Additional research showed that direct-phonon (DP) THz QCL schemes had a clear advantage over resonant-phonon (RP) structures because of their faster LLL depopulation rate [28–31]. An additional advantage of the DP scheme is its low sensitivity to misalignment of the laser levels (caused by the Poisson effect that leads to band bending [28,32,33]). This is achieved because, in a DP scheme, alignment for resonant tunneling is needed only once, whereas resonant tunneling occurs twice in RP schemes.

In this work, we study an SW–DP scheme [34]. Here, the design includes a thin intrawell barrier. By adjusting this barrier, the energy separation between the LLL and the injector level can be tuned to match the exact LO-phonon scattering energy. The excited levels are pushed to higher energies thanks to the intrawell barrier, causing leakage to the continuum and the excited bound states to be suppressed [34]. However, laser operation in these devices only occurs below a $T_{\text{max}}$ of 170 K. Further information about the reasons for this can be found in [35].

To better understand the temperature performance of these devices, we focused on work carried out in 2016 that involved experimental results from three-well RP designs [32]. These results led to the conclusion that high diagonality as a strategy cannot succeed on its own and must be paired with increased doping. $T_{\text{max}}$ was found to improve significantly when the doping was increased from $3 \times 10^{10}$ cm$^{-2}$ to $6 \times 10^{10}$ cm$^{-2}$. The values reported were around 135 K and 177 K, respectively.

Based on this previous work, our expectation was that a similar improvement would occur in other schemes when the carrier concentration was increased from $3 \times 10^{10}$ cm$^{-2}$ to $6 \times 10^{10}$ cm$^{-2}$. Here, we present results showing how doping affects the performance of SW–DP devices. We compared the temperature performance results for a device with a doping concentration of $3 \times 10^{10}$ cm$^{-2}$ with one with double the doping ($6 \times 10^{10}$ cm$^{-2}$). Again, our assumption was that an improvement in $T_{\text{max}}$ would be observed. Ideally the results should have improved but in the devices we checked, the results show the contrary.

In our study we investigated novel diagonal (with oscillator strength of $f \sim 0.2$) SW–DP THz QCLs with mixed potential barriers. The THz QCL scheme contained $Al_{0.55}Ga_{0.45}As$ injection barriers, with the remaining barriers having the standard composition of 15% Al, i.e., $Al_{0.15}Ga_{0.85}As$ (Figure 1 reproduced from Ref. [34]). We realized two devices based on this scheme with different doping levels, namely Device 1 (wafer VB0837) with $\sim 3 \times 10^{10}$ cm$^{-2}$ ($1 \times$ doping) and Device 2 (wafer VB0872) with $\sim 6 \times 10^{10}$ cm$^{-2}$ ($2 \times$ doping). The devices were fabricated and measured together from the same batch. Tables 1 and 2 give details of the device designs, fabrication details, and device parameters. Note that all design parameters were the same for the two devices.
Table 1. Main nominal design parameters and device data.

| Device          | Lasing Energy  | E21 [meV] | Oscillator Strength | Nom. Expected Activation Energy [meV] | E47 [meV] | Layer Sequence [#ML *] | Barrier Composition and Doping Level | Process Details |
|-----------------|----------------|-----------|---------------------|----------------------------------------|-----------|------------------------|--------------------------------------|-----------------|
| Device 1        | 11.1           | 34.5      | 0.26                | 24.9                                   | 72.5      | 9.0/24.8/3.5/24.8/17.3/24.8 | 353 periods                                      | metal–metal     |
| (VB0837)        |                |           |                     |                                        |           | GaAs/mixed barriers     | Al0.55Ga0.45As (Inj.) and Al0.15Ga0.85As (Rad., Intraw.) | (100 Å Ta/2500 Å Au) |
|                 |                |           |                     |                                        |           | in the 24.8 ML wells   | (2.98 × 10^{10} cm^{-2})               | Top contact n+ layer was removed, bottom contact is 50-nm-thick GaAs with doping of 5 × 10^{18} cm^{-3} |
|                 |                |           |                     |                                        |           |                         | Dry etched                           | Mesa size 150 µm x 1.8 mm |
| Device 2        | 11.1           | 34.5      | 0.26                | 24.9                                   | 72.5      | 9.0/24.8/3.5/24.8/17.3/24.8 | 353 periods                                      | metal–metal     |
| (VB0872)        |                |           |                     |                                        |           | GaAs/mixed barriers     | Al0.55Ga0.45As (Inj.) and Al0.15Ga0.85As (Rad., Intraw.) | (100 Å Ta/2500 Å Au) |
|                 |                |           |                     |                                        |           | in the 24.8 ML wells   | (5.96 × 10^{10} cm^{-2}).               | Top contact n+ layer was removed, bottom contact is 50-nm-thick GaAs with doping of 5 × 10^{18} cm^{-3} |
|                 |                |           |                     |                                        |           |                         | Dry etched                           | Mesa size 150 µm x 1.8 mm |

* #ML stands for the number of monolayers, where the AlGaAs barriers are shown in bold, the GaAs wells in roman, and the doped layers in the sequences are underscored. The barriers’ composition and doping details are elaborated in the following text.

Table 2. Device parameters and performance.

| Device (Wafer, Scheme) | Injection Coupling [2hΩij] [meV] | Design Electric Field [kV/cm] | τUL1 [ps] | τUL2 [ps] ** | IFR Gain Broadening [meV] *** | Lasing Energy [meV] | Expected Activation Energy [meV] | f0a (10 K) [A/cm²] | fmax (10 K) [A/cm²] | Dynamic Range (10 K) [A/cm²] | fmax (290 K) [A/cm²] | T
|-----------------------|----------------------------------|-------------------------------|-----------|--------------|-----------------------------|---------------------|---------------------------------|------------------|-----------------|-------------------------------|------------------|---------|
| Device 1 (VB0837)     | 2.08                             | 16.5                          | 1.21      | 0.18         | 4.37                        | 10.05               | 25.5                            | 578              | 928             | 350                           | 750              | 170     |
| Device 2 (VB0872)     | 2.09                             | 16.8                          | 1.32      | 0.18         | 4.37                        | 9.5                 | 26.5                            | 1089             | 1118            | 29                            | 840              | 45      |

* ULL to LLL raw LO-phonon scattering time. ** LLL (level 2) to injector (level 1) LO-phonon scattering time. *** Calculated according to [18].

Figure 1. Band diagram of the SW–DP THz QCLs with mixed barriers: an Al$_{0.55}$Ga$_{0.45}$As injection barrier (nominally a pure AlAs barrier) and Al$_{0.15}$Ga$_{0.85}$As radiative and intrawell barriers. In the figure, two sequential regions define module $i$ (left, marked by dashed-dotted box) and module $i$ + 1 (right), corresponding to the energy levels of Device 1 (VB0837) with a doping level of $\sim 3 \times 10^{10}$ cm$^{-2}$ (1× doping) and Device 2 (VB0872) with double the doping (i.e., $\sim 6 \times 10^{10}$ cm$^{-2}$) (2× doping). Details of design and device parameters can be found in Tables 1 and 2 [34].
2. Discussion

The structure of the devices is shown in Figure 1, where there are three active laser subbands in each module. All other levels seen in the figure are considered parasitic. By means of LO-phonon scattering only, a very fast depopulation rate of the LLL is reached, with no resonant tunneling being involved in the process. The ULL (level 3 in the scheme) and the injector level (level 4 in the scheme) are aligned in the DP scattering scheme. Because of the added barrier of the SW–DP, carrier leakage channels are further reduced, including intermodule leakage.

Pulsed light–current (L–I) measurements for both devices are shown in Figure 2. Increasing the doping of Device 1 (Figure 2a [34]) by a factor of two (i.e., creating Device 2 (Figure 2b)) causes the device performance to degrade substantially, with \(T_{\text{max}}\) being reduced from \(\sim 170\) K to \(\sim 45\) K. The physical mechanism behind the reduced temperature performance observed in the L–I curves is related to the gain broadening accompanying the increased doping [36]. This is a result of the increased overlap of the doped region with the active laser region. The frequency shift between the two devices could be related to band bending. Despite this, from our calculations the difference in \(E_{12}\) is very small and hence negligible.

The I–V curves for Device 1 in Figure 3a [34] show clear NDR behavior at low and room temperatures. The curves become flatter as the temperature increases. The higher the temperature, the lower is the current for the higher-voltage region of the plot. In Figure 3b, Device 2 shows additional flattening of the I–V curve compared with Device 1. Moreover, the decrease in current as the temperature increases, including the value of \(J_{\text{max}}\) (Table 2), is much more severe than that for Device 1. We note from this observation that the \(J_{\text{max}}\) values at room temperature for the 1× and 2× devices are very similar even though the doping is very different.

In Figure 4 the threshold current \(J_{\text{th}}\) is plotted as a function of temperature. It can be observed that \(J_{\text{th}}\) is much higher when the doping is doubled for all temperatures. Although \(J_{\text{th}}\) (Figure 4) increases by a factor of two when the doping is doubled from 1× to 2×, \(J_{\text{max}}\) and the dynamic range \(\Delta J = (J_{\text{max}} - J_{\text{th}})\) scale up by a much lower factor (Table 2). Consequently, \(T_{\text{max}}\) is reduced severely. This is similar to the observations for two-well (TW) structures [28], but with a much faster depopulation rate in the LLL.
Figure 3. I–V curves for: (a) Device 1 (VB0837, 1× doping) [34] and (b) Device 2 (VB0872, 2× doping) at low and room temperatures. $T_{\text{max}}$ values are also shown.

Figure 4. Plots of the SW–DP THz QCL devices’ threshold current versus temperature.

In a clean three-level device, the $J_{\text{max}}$ versus temperature relationship carries important information that might explain how different mechanisms affect the temperature performance. The Kazarinov–Suris [34,37] expression for current tunneling describes transport through the injector barrier:

$$J = eN \times \frac{2\Omega^2 \tau_{\parallel}}{4\Omega^2 \tau_{\parallel} + \omega_{21}^2 \tau_{\parallel}^2 + 1}$$  \hspace{1cm} (1)

where $\Omega$ is the coupling between the injector and the ULL subbands across the barrier, $\tau$ is the ULL lifetime, $\tau_{\parallel}$ is the decoherence (dephasing) time between the injector and the ULL subbands, and $\omega_{21}$ is the energy misalignment between the two. Equation (1) describes a Lorentzian function centered at $\omega_{21} = 0$, whose width is proportional to $\sqrt{4\Omega^2 \tau_{\parallel} + 1}$. The maximum current ($J_{\text{max}}$) is then given by $J(\omega_{21} = 0) = J_{\text{max}}$. A weak coupling regime (resonant-tunneling-limited transport) corresponds to the case where $\tau \ll \frac{1}{4\Omega^2 \tau_{\parallel}}$, whereas a strong coupling regime (lifetime-limited transport) corresponds to the case where $\tau \gg \frac{1}{4\Omega^2 \tau_{\parallel}}$ [38,39]. Diagonal design follows the latter regime. Here, because of the reduction in the ULL lifetime upon lasing, the transport can be affected by stimulated emission. Consequently, the maximum current density prior to the onset of NDR (i.e., $J_{\text{max}}$)
may decrease with temperature if the transport is strongly limited by the ULL lifetime (i.e., the strong coupling regime), as for diagonal designs, because $J_{\text{max}} \sim \frac{1}{\tau} \approx \frac{1}{\tau_{\text{st}}}$. It follows from this that the dynamic range is reduced as the temperature rises. Because $J_{\text{max}} \sim \frac{1}{\tau} \approx \frac{1}{\tau_{\text{nr}}}$, the maximum current will change direction and start to escalate when lasing stops at $T_{\text{max}}$. The nonradiative lifetime $\tau_{\text{nr}}$ will now dominate the ULL lifetime and will decrease as the temperature increases. This behavior continues until the current is limited by resonant tunneling (i.e., a transition to the weak coupling regime). At this point, $J_{\text{max}}$ will continue to reduce because now $J_{\text{max}} \sim \tau_{\text{dl}}$ and the decoherence time between the injector and ULL (i.e., $\tau_{\text{dl}}$) declines as the temperature increases. The identification and analysis of these three regions—namely Region 1: strong coupling regime when lasing ($T \leq T_{\text{max}}$), Region 2: strong coupling regime when not lasing ($T > T_{\text{max}}$), and Region 3: the weak coupling regime—are crucial to understanding the behavior of our scheme.

The plot of the maximum current $J_{\text{max}}$ versus temperature in Figure 5 shows these three regions for our two devices, together with their $T_{\text{max}}$ values. Two main observations can be made regarding the impact of doping in our devices. First, the impact of doping is clear from the comparison of the maximum current $J_{\text{max}}$ versus temperature data for Device 1 and Device 2. Device 2 demonstrates an earlier transition from the strong coupling regime (Regions 1 and 2) to the weak coupling regime (Region 3), which indicates the negative effect of doping on the dephasing time. Second, when the temperature rises, line broadening increases. Line broadening eventually exceeds energy splitting, meaning that transport is controlled by incoherent tunneling and the wave functions become localized.
show a faster drop in the maximum current for Device 2. Eventually, at room temperature, the maximum currents for Device 1 and Device 2 are very similar. The faster drop of $J_{\text{max}}$ indicates a faster dephasing rate as the temperature increases, making the degree of wave-function localization significant at room temperature.

Because the IFR effects are similar for the two devices, the main mechanism that affects the dephasing with increase of doping is the increase of line broadening caused by ionized impurity scattering (IIS). This becomes more significant as the doping level increases. These results are consistent with earlier theoretical works \cite{21,22}. Our work has therefore demonstrated experimentally effects related to dephasing that to date were accessible only via simulation \cite{21–23}.

In conclusion, our unique designs have allowed us to observe experimentally the effect of doping on dephasing times and line broadening. Although an increase in dopants has a positive effect for RP schemes, our results showed that this does not occur in SW–DP devices. We observed a significant increase in line broadening as the doping and temperature increased and attribute this effect to enhanced IIS. These insights into the role of doping suggest an approach to improving THz QCLs by engineering the doping profile and its spatial location. Applying this approach in future should involve carefully engineering the devices to minimize the overlap between the doped region and the active level region. The effects of IFR scattering should be explored with similar SW–DP variable-barrier structures. Our approach should serve as an excellent platform for eventually achieving terahertz frequency lasing at room temperature.

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