Correlation of Superlattice Cross-Plane Thermal Conductivity with Emission Wavelength in InAlAs/InGaAs Quantum Cascade Lasers

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ABSTRACT

The low cross-plane thermal conductivity of quantum cascade lasers (QCLs) is a significant limitation in their Continuous-Wave (CW) performance. Structural parameters such as individual layer thicknesses and interface density vary for QCLs with different target emission wavelengths, and these design parameters are expected to influence the cross-plane thermal conductivity. Though previous works have used theoretical models and experimental data to quantify thermal conductivity, the correlation between target wavelength and thermal conductivity has yet to be reported for QCLs. In this work, we observe a general trend across a group of QCLs emitting from 3.7 to 8.7 μm: as the QCL design changes to reduce wavelength, the thermal conductivity decreases as well. Numerically, we measured an approximate 70% reduction in thermal conductivity, from 1.5 W/(m·K) for the 8.7 μm device, to 0.9 W/(m·K) for the 3.7 μm device. Analysis of these structures with the Diffuse Mismatch Model (DMM) for thermal boundary resistance (TBR) shows that the largest contribution of this effect is the impact of superlattice interface density on the thermal conductivity. The observed changes in conductivity result in significant changes in projected CW optical power and should be considered in laser design.

1. INTRODUCTION

Quantum Cascade Lasers (QCLs) are semiconductor laser devices that generate infrared light through inter-sub-band laser transitions. QCLs are composed of alternating nanometer-thick semiconductor layers in a superlattice structure that generate photons as an electron cascades through the quantum well structure. The active region is composed of alternating layers of ternary semiconductor materials which serve as the barrier and well of the quantum cascade structure. QCL design hinges on bandgap engineering to create high power tunable infrared laser light that is guided within the active medium. QCL advancements have allowed for room-temperature Continuous Wave (CW) operation of QCLs, but a persistent limitation in the widespread application of QCLs is the problem of thermal dissipation in the superlattice laser core1-4. This limitation can mainly be attributed to the density of individual ternary semiconductor layers that make up the superlattice1-5, as well as the high power density typical of QCLs1-6. This limitation necessitates that QCL active region design considers the effective thermal conductivity of the core, which significantly impacts heat dissipation. Thermal conductivity within the active region is anisotropic, and there is a considerable discrepancy in the ability of the core to dissipate heat in the lateral direction (towards the sidewalls) as compared to the cross-plane direction, a direct consequence of the superlattice interfaces1.

State of the art QCL superlattices are typically composed of InAlAs barriers and InGaAs quantum wells4-10, with parameters such as the number of pairs and the individual thicknesses varying across different QCL structures depending on the target wavelength. To achieve lower and lower target wavelengths, the QCL active region must be modified. Specifically, the total quantum well thickness reduces while barrier thickness increases to accommodate a larger energy spacing between upper and lower laser level. The well material has the lower conductivity, and so the weighted average of thermal conductivity is expected to reduce as well. Additionally, shorter wavelength lasers require a larger number of well/barrier pairs to achieve the same voltage defect with thinner wells. This increase in the number of interfaces is detrimental to the thermal conductivity in this critical cross-plane direction. The individual contributions from a reduced target wavelength do lend themselves to a general reduction in target wavelength, but they alone do not guarantee a trend. These modifications do result in a marked increase in the superlattice interface density, which in turn hinders heat transport.
and causes the most significant reduction in cross-plane thermal conductivity\(^5\). In this work we aim to quantify the correlation between target emission wavelength and cross-plane thermal conductivity in a series of QCLs that encompass a range of wavelengths from 3.7-8.7\(\mu m\).

The thermal transport problem in QCLs has received a lot of attention in literature\(^1,2,4-9,12\). Previously, various theoretical approaches have been used for determining key thermal properties, and many experimental approaches have been used for probing the temperature profiles QCLs\(^13-16\). Through adaptations of these methods, we probe the relationship between thermal conductivity and wavelength, which to our knowledge has been unexplored in the wavelength regime we are considering here (Mid-Infrared to Long Wave Infrared). This is accomplished through an experimental approach as in Refs. 15 and 16, as well as through an adaptation of the theoretical model described in Refs. 3 and 13. In both cases, we can confirm the anticipated trend, as well as quantify the significance of the interface density on this trend. Lastly, we demonstrate how this reduced thermal conductivity affects CW performance.

2. MATERIALS AND METHODS

The lasers considered were all InP-based buried heterostructure lasers. These chips were made through the regrowth of Iron-doped InP on the sidewalls of a ridge waveguide. Four different laser core designs were considered, and each given a designation for identification. The S12 laser structure emitted in the LWIR regime at 8.7\(\mu m\). The remaining laser devices were all in the MID range. The S1 structure emitted at 5.7\(\mu m\), and the remaining two lasers (S25 and D41) both emitted at 3.7\(\mu m\). The two 3.7\(\mu m\) devices had slightly varying design. For each structure, laser chips were mounted epide side down onto ceramic AlN sub-mounts and coated with a highly reflective coating. The lasers were tested in CW at 15 °C using a chiller to maintain heat sink temperature. A theoretical phonon transport model based on some basic material parameters was used to theoretically determine thermal conductivity for comparison with the experimental results. After Light-Current-Voltage (LIV) curves were measured, the laser temperature characteristics were determined using a micro-Raman thermometry setup paired with COMSOL thermal simulations to extract the cross-plane thermal conductivity for each laser.

2.1 Theoretical Approach

Previous works in the realm of THz QCLs used a theoretical phonon transport model for semiconductor superlattices\(^2\) to project changes in thermal conductivity. This model factors in the main obstacle to heat dissipation in superlattices: the countless individual interfaces that impede heat transfer. This affects the cross-plane conductivity by way of the Thermal Boundary Resistance term (TBR), which quantifies the heat flow between two solid interfaces of different materials\(^17-19\). The anisotropic thermal conductivity is calculated separately for the in-plane and cross-plane direction. While the in-plane thermal conductivity does not consider TBR since there are no superlattice interfaces and is therefore a simple series summation, the cross-plane thermal conductivity requires parallel summation with the added term that represents the overall thermal resistance contributed by the superlattice interfaces\(^3,13,19\). The thermal conductivity is calculated as follows:

\[
\begin{align*}
    k_\parallel &= \frac{L_1}{L} k_1 + \frac{L_2}{L} k_2 \\
    \kappa_\perp^{-1} &= \frac{L_1}{L} k_1^{-1} + \frac{L_2}{L} k_2^{-1} + \frac{N}{L} \ast R^{(ave)}
\end{align*}
\]

where \(L_i\) is the total thickness of the \(i_{th}\) material in a single stage of the superlattice, \(k_i\) is the thermal conductivity of the \(i_{th}\) material in the superlattice, \(L\) is the total thickness of a single stage, \(N\) is the number of interfaces in a stage, and \(R^{(ave)}\) is the average of the thermal boundary resistance of heat flow in both directions of the interface\(^2\). TBR is calculated from the material properties of the constituent superlattice materials\(^6,17-19\), InGaAs and InAlAs, and its contribution to the thermal conductivity is weighted based on the density of interfaces in a single stage, \(N/L\). The relevant parameters needed for the calculation of TBR are given in Table 1.
Table 1. Material parameters of samples in this study

| Structure | Wavelength (µm) | Number of Interfaces | Total Stage Thickness (Å) | Laser Core Dimensions | Barrier Composition/Total Thickness (Å) | Quantum Well Composition/Total Thickness (Å) |
|-----------|-----------------|----------------------|--------------------------|-----------------------|----------------------------------------|-------------------------------------------|
| D41       | 3.7             | 20                   | 427                      | 5 mm × 8 µm           | Al$_{0.78}$In$_{0.22}$As                | Ga$_{0.24}$In$_{0.76}$As                 |
|           |                 |                      |                          |                       | 188                                    | 239                                      |
| S25       | 3.7             | 20                   | 447                      | 5 mm × 7.2 µm         | Al$_{0.78}$In$_{0.22}$As                | Ga$_{0.23}$In$_{0.77}$As                 |
|           |                 |                      |                          |                       | 198                                    | 249                                      |
| S1        | 5.7             | 18                   | 453                      | 3.15 mm × 7 µm        | Al$_{0.78}$In$_{0.22}$As                | Ga$_{0.37}$In$_{0.63}$As                 |
|           |                 |                      |                          |                       | 160                                    | 293                                      |
| S12       | 8.7             | 16                   | 449                      | 5 mm × 9 µm           | Al$_{0.65}$In$_{0.35}$As                | Ga$_{0.41}$In$_{0.59}$As                 |
|           |                 |                      |                          |                       | 115                                    | 334                                      |

These parameters are used to calculate TBR but are also input parameters in the COMSOL thermal simulation, specifically the laser core dimensions and total stage thickness.

The TBR is taken to be the average of the TBRs for both directions of heat flow. The TBR, and subsequently the thermal conductivity, were calculated at room temperature.

\[
R^{(\text{ave})} = \frac{R_{1\rightarrow2} + R_{2\rightarrow1}}{2}
\]  

Here \( R_{i\rightarrow j} \) is the TBR from material \( i \) to \( j \) in units of m$^2$K/W.

\[
R_{i\rightarrow j} = \left[ \frac{1}{2} \sum_j v_n \Gamma_{i,j} \int_0^{\omega_D} \frac{\hbar \omega}{dN_{i,j}(\omega,T)} d\omega \right]^{-1}
\]

where \( v_{n,j} \) denotes the acoustic phonon velocity. The subscript \( n \) denotes the material number, while the subscript \( j \) denotes the polarization direction (1 being the longitudinal direction, while 2 and 3 being transverse. \( \omega_D \) is the Debye frequency, \( N_{n,j} \) is the density of phonon states, and \( \Gamma_{n,j} \) is the averaged transmission coefficient. These last two values are defined as:

\[
N_{1,j}(\omega,T) = \frac{\omega^2}{2\pi^2 v_{1,j}^3 \left[ \exp \left( \frac{\hbar \omega}{k_B T} \right) - 1 \right]}
\]

\[
\Gamma_{1,j} = \int_0^\pi \alpha_{1\rightarrow j}(\theta, j) \cos \theta \sin \theta \, d\theta
\]

The average transmission coefficient is dependent on the transmission probability term. The calculation of this term can be done through one of two ways based on the approach taken to model the phonon transport across the interfaces. Two models exist, the Acoustic Mismatch Model (AMM) and the Diffuse Mismatch Model (DMM). The AMM considers the polarization state of the phonons and treats them as elastic waves passing through continuous media separated by an ideal plane\cite{2,18,19}. In addition to requiring the material acoustic phonon velocity, the transmission probability requires the density \( \rho_n \) of each material. The DMM model considers the effect of phonon scattering at the interface and assumes that the phonons that traverse the interface scatter diffusively, losing all coherence, as well as any information on their previous polarization states\cite{17,18}. These two models represent two extrema of possibility for phonons crossing the interface. Ref 2 points out that although phonon scattering processes can be more accurately described by an average of the two model approaches, weighed off of a specularity parameter that is defined by material parameters such as roughness, considering the TBR purely based off one of the two models is sufficient for estimation of the cross-plane thermal conductivity. The transmission probability for each model is defined as:

\[
\alpha_{1\rightarrow 2,\text{DMM}} = \frac{\sum_j v_{n,j}^2 \Gamma_{1,j}}{\sum_{n,j} v_{n,j}^2 \Gamma_{n,j}}, \quad \alpha_{1\rightarrow 2,\text{AMM}} = \frac{A\rho_1 v_{1,j} v_{2,j} \cos \theta_1 \cos \theta_2 \cos \theta_2}{(\rho_1 v_1 \cos \theta_1 + \rho_2 v_2 \cos \theta_2)^2}
\]
Where \( \theta_n \) is the incident phonon angle based on Snell’s law at the interface. We can see that because the DMM does not consider the carry-over of polarization states, the transmission probability is independent of said incident angle\(^2\). Table 2 illustrates the relevant parameters for calculating TBR as they depend on the Ga/Al distribution of the two superlattice constituents.

Table 2. Composition-dependent material parameters for TBR Analysis

| Parameter                              | Material 1 \((Al_xIn_{1-x}As)\)\(^a\) | Material 2 \((Ga_xIn_{1-x}As)\)\(^b\) |
|----------------------------------------|--------------------------------------|--------------------------------------|
| Elastic Constant \(C_{11}\) \(\frac{N}{m^2}\) | \((8.34 + 3.68x) \times 10^{10}\)   | \((8.34 + 3.56x) \times 10^{10}\)   |
| Elastic Constant \(C_{44}\) \(\frac{N}{m^2}\) | \((3.95 + 1.94x) \times 10^{10}\)   | \((3.95 + 2.01x) \times 10^{10}\)   |
| Material Density \(\rho\) \(\frac{kg}{m^3}\) | \(5680 – 1920x\)   | \(5680 – 370x\)   |
| Longitudinal Acoustic Wave Speed \(v_L\) \(\frac{m}{s}\) | \(\sqrt{\frac{C_{11}}{\rho}}\)       | \(\sqrt{\frac{C_{44}}{\rho}}\)       |
| Transverse Acoustic Wave Speed \(v_T\) \(\frac{m}{s}\) | \(\sqrt{\frac{C_{44}}{\rho}}\)       | \(\sqrt{\frac{C_{44}}{\rho}}\)       |
| Debye Temperature \(\Theta_D\) [K]     | \(280 + 166x\)   | \(280 + 110x\)   |
| Bulk Thermal Conductivity \(k\) \(\frac{W}{mK}\) | \(2.5\)   | \(5.0\)   |

\(a\) Interpolated from material parameters of InAs, InAlAs and AlAs taken from Ref. 6,8,11,20. \(b\) Taken from Ref. 6,8,11,20.

### 2.2 Experimental Approach

Firstly, the CW LIV curves of each device were measured at 15°C heat sink temperature. Following this, the impact of wavelength selection on thermal conductivity was experimentally determined across 4 lasers. Raman thermometry\(^15,16\) was used to locally measure the output facet temperature of each QCL as a function of the injected current in CW. The same approach as in Ref. 15 was used. Firstly 532 nm laser light was focused with a 50X objective lens onto the front facet. A spot size of \(~1.3\) \(\mu m\) was illuminated with 0.6mW of optical power. The backscattered light was measured by the spectrometer and was first used to determine the linear relationship between Raman peak frequency and temperature. This was determined to be:

\[
T = T_0 - (\omega_0 - \omega_m)m
\]

Where \(\omega_0\) is the room temperature \((T_0)\) Stokes peak position, \(m\) is the slope of the linear relationship, and \(\omega_m\) is the Stokes frequency peak position measured from the device. The laser was then connected to a CW current source, and the Raman spectra was measured in increments of 0.1 A up to laser rollover. 6 spectra were taken at each current and the averaged Raman shift peak position was used to determine the temperature at the facet as a function of injection current.

The experimental cross-plane thermal conductivity was determined by simulating the core temperature of each device as a function of CW electrical power and comparing these results to the experimental curves, with thermal conductivity as the sole fitting parameter. The COMSOL Multiphysics heat transfer model was used to project joule heating from the input electrical power while also considering the temperature dependence of the thermal conductivity. The room-temperature thermal conductivity was taken from the best fit of the simulated temperature curves with the experimental data, as in Ref 15. Lastly, a 2D thermal simulation which extracts the temperature vs current curves as a function of thermal conductivity was used to project the CW power of the S25 laser. The resulting CW LI curve was compared to the experimental LI data to explore the impact the conductivity has on laser performance\(^21\).

### 3. RESULTS

### 3.1 Theoretical Results

Using the parameters in Table 2 we were able to calculate the thermal boundary resistance and thermal conductivity for each laser based on both the DMM and AMM. Additionally, the interface density was measured in each case to confirm its dependence on the wavelength. The results are shown in Table 3.
Table 3. Results of the calculation of cross-plane thermal conductivity and TBR using the DMM and AMM

| Wavelength (µm) (Structure) | Interface Density N/L (nm⁻¹) | DMM Thermal Boundary Resistance (m²K/W) | AMM Thermal Boundary Resistance (m²K/W) | DMM Cross-Plane Thermal Conductivity (W/m/K) | AMM Cross-Plane Thermal Conductivity (W/m/K) |
|-----------------------------|-------------------------------|----------------------------------------|----------------------------------------|---------------------------------------------|---------------------------------------------|
| 3.7 (D41)                  | 0.468                         | 1.549 × 10⁻⁹                         | 9.765 × 10⁻⁹                          | 0.986                                       | 1.341                                       |
| 3.7 (S25)                  | 0.447                         | 1.557 × 10⁻⁹                         | 9.83 × 10⁻⁹                           | 1.015                                       | 1.373                                       |
| 5.7 (S1)                   | 0.397                         | 1.452 × 10⁻⁹                         | 8.9 × 10⁻⁹                            | 1.18                                        | 1.602                                       |
| 8.7 (S12)                  | 0.356                         | 1.445 × 10⁻⁹                         | 8.4 × 10⁻⁹                            | 1.305                                       | 1.816                                       |

Thermal conductivity decreases with shorter wavelengths according to both theoretical models. Additionally, we show that the interface density follows an inverse trend, as it gets higher at shorter wavelengths. As will be shown in the experimental discussion, the DMM model more closely matches the experimental results for thermal conductivity, falling within 10-15% of the results, while the AMM appears to overestimate the conductivity for these lasers. The DMM model is more applicable in the case where high interface roughness causes diffuse scattering to dominate²⁻²⁰, and this is expected to be the case in these lasers, with some exception in the long-wave regime that will be discussed in the experimental results. Notably, the TBR for D41 in both models is lower than that of S25, which indicates that the TBR is not the deciding factor in the correlation between thermal conductivity and wavelength. Rather, the interface density, which acts as a weighing factor for the TBR, serves as the prime contributor to the thermal conductivity in the cross-plane direction, and results in D41 having a 3% lower thermal conductivity compared to S25. The relative importance of this term can be understood by breaking down each term of Eq 2. Table 4 Summarizes these terms.

Table 4. Values for each term in cross-plane thermal conductivity calculation

| Wavelength (µm) | \( \frac{L_1}{L} k_1^{-1}[\frac{mK}{W}] \) | \( \frac{L_2}{L} k_2^{-1}[\frac{mK}{W}] \) | \( \frac{N}{L} R^{(ave)}[\frac{mK}{W}] \) |
|----------------|------------------------------------------|------------------------------------------|------------------------------------------|
| 3.7            | 0.176                                    | 0.112                                    | 0.726                                    |
| 3.7            | 0.177                                    | 0.111                                    | 0.697                                    |
| 5.7            | 0.141                                    | 0.13                                     | 0.577                                    |
| 8.7            | 0.102                                    | 0.149                                    | 0.515                                    |

The dominant term in the calculation is the thermal boundary resistance, weighed based on the density of interfaces. The relationship between interface density and cross-plane thermal conductivity is shown in Figure 1.
Figure 1. Theoretical relationship between interface density and thermal conductivity as per the DMM. Error bars representing the uncertainty of the fit.

Figure 2. Temperature curves for the S12 device emitting at 8.7 μm. Cross-plane thermal conductivity is reported as 1.5 W/(m·K). Device dimensions are 5 mm × 9 μm.

The S12 device is the only LWIR among the five tested. LWIR devices can suffer additional heating from optical reabsorption due to facet oxidization. This device was coated and tested immediately in order to avoid excessive exposure to the ambient environment that would cause oxidization. Therefore, no change in heating rate above threshold was seen. This laser, being the longest wavelength, also had the highest thermal conductivity. Notably this device showed the greatest deviation from the predicted thermal conductivity, suggesting its interfacial phonon transport processes are not properly described by just the DMM.
Figure 3. Temperature curves for the S1 device emitting at 5.7 μm. Cross-plane thermal conductivity is reported as 1.2 W/(m·K). Device dimensions are 3.15 mm × 7 μm.

This laser uses an active region design with record efficiency10, and it also showed the greatest adherence of the thermal conductivity to the predicted model result.

Figure 4. Temperature curves for S25 device emitting at 3.7 μm. Cross-plane thermal conductivity is reported as 0.9 W/(m·K). Device dimensions are 5 mm × 7.2 μm.
The last two devices both emitted at the same wavelength, but had variation in active region parameters as seen in Table 1. These two lasers had roughly the same thermal conductivity, although the model more accurately reported S25 as having a slightly higher cross-plane thermal conductivity. For both devices, the thermal conductivity was measured to be less than the DMM prediction. Figure 6 summarizes the experimental cross-plane conductivity, as well as the two model results, for each laser wavelength tested.

Just as in the theoretical case, the cross-plane thermal conductivity decreases as wavelength is reduced. In all five lasers, the experimental data most closely matches the DMM thermal conductivity. A notable deviation is the experimental S12 conductivity, which is ~10% higher than the DMM prediction. This suggests that the S12 laser has a lower interface roughness than the lasers in the MIR range, and the interfacial scattering phenomenon is instead best represented by a combination of the two models.\(^2,20\).
4. DISCUSSION

This correlation cannot be ignored in laser design considerations. To illustrate the impact of the conductivity, we modeled the CW LI curves of the S25 at two different extrema of cross-plane thermal conductivity, from 0.9 to 1.5 W/(m·K). A 2D thermal model of the S25 structure was used alongside a numerical model described in Ref. 21 to project the CW power of this laser. Figure 7 shows the results of this analysis. The CW power projected for the experimental cross-plane conductivity was in good agreement with the experimental LI curve, while the power projection taken from the upper extreme of conductivity overestimated the maximum power by 30%. It is expected that this difference would be even higher for wide active region devices as thermal conductivity is more important for heat dissipation in these cases.

![Figure 7. CW Power projection for the two extremes of thermal conductivity for the S25 structure.](image)

5. CONCLUSION

Over a range of QCL wavelengths, we have demonstrated a correlation between emission wavelength and cross-plane thermal conductivity. This is confirmed not only experimentally, but with two different models for thermal conductivity. From these models, we probed the significance of each superlattice parameter, and determined that the density of interfaces in a single stage, which increases at lower wavelengths, is the limiting factor for thermal conductivity. We learned that all the MIR structures (<8 μm), adhere most to the DMM prediction, which suggests the interfacial roughness of these devices is considerable enough for diffusive scattering to dominate. The LWIR device, S12, likely has less interface roughness, and therefore experimentally it falls between the value given by both models. Probing the quality of interface roughness of these QCLs would allow one to more accurately determine thermal conductivity based on TBR.

This previously unexplored relationship lends some significant insight into the design of QCLs, and the CW projection at the two wavelengths demonstrates how much of an impact the conductivity has on power output. Reducing the interface density in these low wavelength devices would allow for better device performance. The two structures of the same wavelength had slightly different interface densities, which shows that one can modify the QCL structure while maintaining the same target emission wavelength. Emphasizing this goal in QCL design could lead to marked improvement in laser performance for shorter QCL lasers.

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