Importance of coherence in models of mid-infrared quantum cascade laser gain spectra

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Abstract

We present a three-level model based on a density matrix to examine the influence of coherence and dephasing on the gain spectrum of mid-infrared quantum cascade lasers. The model is used to examine a quantum cascade active region with multiple optical transitions. We show how coherence can explain the origin of additional peaks in the gain spectrum. We also analyze the spectra calculated using the three-level model with a rate equation formalism to demonstrate the importance of considering interface roughness and limitations of the rate equation formalism. Specifically, we present how interface roughness influences the broadening and oscillator strength that are recovered using a rate equation analysis. The results of this work are important when considering the design of active regions with multiple optical transitions and could lead to devices with improved performance.

Keywords: quantum cascade lasers, density matrix, interface roughness scattering

1. Introduction

Engineered bound states in quantum wells are the heart of quantum cascade lasers (QCLs), and proper understanding of these quantum states—both individually and collectively—has enabled rapid evolution in the performance of QCLs. Since the first demonstration in 1994 [1], QCLs with remarkable performance have been realized, including: continuous wave operation at room temperature [2]; high electrical-to-optical conversion efficiency [3–8]; and, nonlinear generation of mid-infrared [9] and THz radiation [10]. These advances, and many others, are partly the result of improved understanding of intersubband physics and clever quantum engineering of the conduction band profile. High-performance QCLs have since been demonstrated over much of the mid-infrared region [8], enabling applications such as spectroscopy and chemical sensing [11, 12].

The designed conduction band influences nearly all aspects of the operating characteristics of QCLs. While the active regions are unique, lasers designed for broad gain or difference frequency generation share some characteristic design features. In particular, many of the designs for these QCLs include several nearly resonant energy levels with large dipole matrix elements. For broad gain QCLs, these levels broaden the gain spectrum by providing multiple optical transitions [13–15], and for nonlinear QCLs, the levels result in a large nonlinear susceptibility [9, 10]. For both types of these QCLs, one strategy to realizing nearly resonant levels is to design a heterostructure that supports delocalized single electron wavefunctions that span many wells and barriers.

Rate equation models for such design approaches may have
limitations due to the small energy difference between eigenstates and the relatively large energy broadening.

Recent progress in the wall-plug efficiency of QCLs addresses energy broadening due to interface roughness. This prior research demonstrates the importance of considering physics beyond conventional rate equation models and considers effects such as coherence and dephasing in QCL heterostructures. In rate equation models, electron transitions are described as scattering processes with an effective scattering time; however, coherent processes cannot be treated as an effective scattering process because the phase of the wavefunctions influences the transition. Such coherent processes are readily described using a density matrix model. The influence of coupling between the injector and upper-laser level (u.l.) was examined using a three-level model based on a density matrix [16]. It was shown that for typical QCL designs, the spectral broadening of the primary optical transition is determined primarily by interface roughness scattering. It was also shown that increasing the coupling strength between the injector level (i.l.) and the u.l. would increase the peak gain with minimal broadening of the spectrum. The importance of this model was captured in the demonstration of so-called ‘ultra-strong coupling’ QCLs. These devices incorporated stronger coupling between the i.l. and u.l. and exhibited wall-plug efficiencies of 50% [5]. The simple, but insightful, three-level model based on a density matrix enabled new insight into QCL design and significant gains in performance. In the terahertz region, density matrix models have also been developed to better understand the influence of coherence on the gain profile [17]. Density matrix models that incorporate more than three levels have also been developed to aid in the understanding and design of QCL active regions [18–21]. These models are often evaluated numerically and while insightful, developing an intuition for QCL design from such complicated models is difficult.

In this paper, we present a three-level density matrix model that gives insight into QCL active regions that incorporate multiple optical transitions. We apply this model to a QC heterostructure that is similar to one of the structures presented in [22] (sample D2159). This particular structure is of interest because the lineshape of the measured electroluminescence is a Lorentzian peak with a low-energy shoulder, and this simple emission spectrum can be used to demonstrate the effects of coherent coupling and dephasing on the gain spectrum. We demonstrate how the density matrix model can be used to understand QCL active regions with nearly resonant energy levels. Furthermore, we show how using conventional rate equations can lead to poor estimations and understandings of active region parameters such as the dipole matrix element and broadening.

2. Model description

Figure 1 depicts the conduction band for a single period of the modeled structure comprising InGaAs wells and AlInAs barriers. Each period of the structure consists of one active region and one injector, and we define the extractor region as the injector of the next period, figure 1. The thicknesses of the AlInAs/InGaAs layers in nm for the structure are 4.5/8.0/1.0/5.7/1.8/4.4/1.4/3.6/1.2/3.6/1.2/3.4/1.0/3.4; where the barriers are in bold. For each wavefunction, \( \psi_n \), we plot \( |\psi_n|^2 \) at the energy that corresponds to the calculated eigenenergy, \( E_n \). Four single electron wavefunctions of particular interest in gain calculations are labeled from 0 to 4 in figure 1. These wavefunctions correspond to the i.l. (0), the u.l. (1), the lower-lasing level (l.l.) (2), and the extractor level (e.l.) (3); the e.l. is the level similar in energy to level 2 but located mainly in the downstream injector. The subband states shown in figure 1 are non-stationary solutions to the Hamiltonian for the conduction band potential. When states are close in energy, the non-stationary wavefunctions are more localized in space compared to the often-calculated stationary wavefunctions. In a model describing electron transport and photon emission, non-stationary states are favored because coherent coupling between them is important.

The need to account for coherence and dephasing is evident when considering the optical characteristics of multi-peaked gain spectra; for example, the electroluminescence of a device with the design shown in figure 1. An analysis of this spectrum, which exhibits a low-energy shoulder on a larger emission peak [22], using rate equations is complicated due to the unknown populations of the subbands. As such, it is difficult to assign the low-energy shoulder to the 0 \( \rightarrow \) 2 or 1 \( \rightarrow \) 3 transition (or a combination of the two), complicating further engineering or optimization of the active region. Additionally, a rate equation approach does not properly treat energy broadening from such diagonal transitions.

An alternative approach to understanding the emission spectrum is to employ a density matrix model. We use two three-level density matrix models to calculate the gain lineshape of the quantum well heterostructure in figure 1. The first model was developed in [16] and involves levels 0, 1, and 2. This model includes coherent coupling between the i.l. and u.l. and is referred to as the INJ model. The second
model, developed in this paper, comprises levels 1, 2, and 3; this model includes coherent coupling between the l.l. and the e.l. and is referred to as the XTR model. In mid-IR QCLs, longitudinal optical (LO) phonon scattering is usually the key process that limits mobility [23], while interface roughness scattering dominates the broadening of the optical linewidth [24]. In the density matrix models, we include LO phonon scattering and interface roughness scattering and ignore all other scattering sources. The following sections describe how we model coherent coupling between states and scattering.

2.1. Coherent coupling

The coupling energy describes coherent oscillation of the electron population between states separated by a potential barrier, such as the l.l. and e.l. This oscillation occurs at the Rabi oscillation frequency. For both models, the coupling energy between two wavefunctions, \( \psi_a(z) \) and \( \psi_b(z) \), is determined via the method in [25]:

\[
\int_{-\infty}^{\infty} dz \psi_b(z) \Delta V(z) \psi_b(z),
\]

where \( z \) is the spatial position along the growth axis and \( \Delta V(z) \) is a perturbation that accounts for coupling between neighboring isolated periods of the QCL active region, akin to a tight-binding model. For the heterostructure studied here, and most QCLs in general, the coupling energy between levels 0 and 1, \( \hbar \Omega_{101} \approx 2 \text{ meV} \), is smaller than the coupling energy between levels 2 and 3, \( \hbar \Omega_{32} \approx 6 \text{ meV} \), primarily due to the fact that the injection barrier is thicker than the extraction barrier.

2.2. Interface roughness

Dephasing refers to processes that destroy the coherent superposition between two states, ultimately broadening the transition energy. The transition broadening due to interface roughness scattering is largely influenced by the intrasubband contribution due to the difference in the scattering matrix elements [23]. In a Gaussian roughness model, interface roughness is characterized by an average height \( \Delta \) and correlation length \( \Lambda \). The broadening between two states, \( \psi_m(z_j) \) and \( \psi_n(z_j) \), due to interface roughness scattering is given by [16, 26]:

\[
\hbar \tau^{-1}_m = \frac{m^*}{\hbar} \Delta^2 \Lambda^2 \delta U^2 \sum_j \left| \psi_m^2(z_j) - \psi_n^2(z_j) \right|^2,
\]

where the sum is over each interface at \( z_j \), \( \hbar \) is the reduced Planck constant, \( m^* \) is the electron effective mass, and \( \delta U \) is the conduction band offset.

2.3. LO phonon scattering

Rapid LO phonon scattering contributes to the overall decoherence via relaxation. The LO phonon scattering rates are calculated for each level; these rates for the XTR model are indicated in figure 2 as \( \tau_1 \), \( \tau_2 \), and \( \tau_3 \) for levels 1, 2, and 3, respectively. Typically \( \tau_2 \) and \( \tau_3 \) are dominated by the levels immediately below levels 2 and 3 (see figure 1) so that fast scattering of electrons from these two levels is ensured. To capture this, we use a forth auxiliary level from the complete bandstructure to calculate \( \tau_2 \) and \( \tau_3 \); both are about 0.5 ps. Scattering from the auxiliary level to the next period’s level 1 is assumed to be instantaneous. In the XTR model, the actual electron populations are not correct, since the approach does not properly treat injection into level 1. However, the purpose of this model is not to calculate the value of the gain, but rather the impact of coherence and dephasing on the lineshape and the gain spectrum—the model is appropriate for this purpose.

The INJ and XTR models employ similar quantum mechanical descriptions of coherent coupling, dephasing due to interface roughness, and phonon assisted scattering. An energy level diagram for the XTR model is shown in figure 2. The primary optical transition is between levels 1 and 2, the u.l. and l.l., respectively. The designed emission energy is \( h\omega_{12} = 143 \text{ meV} \). Detuning between the photon field frequency, \( \omega \), and the primary optical transition is given by \( \Delta = \omega_{12} - \omega \). State 3, the e.l., is nearly resonant with state 2. The calculated coupling energy is \( \hbar \Omega_{32} = 6.3 \text{ meV} \), which is nearly field-independent around the operating field of the device. The two levels are detuned in energy by \( \hbar \delta \). Each pair of levels has an associated energy broadening due to interface roughness scattering that are indicated as \( \hbar \tau^{-1}_{12}, \hbar \tau^{-1}_{23}, \) and \( \hbar \tau^{-1}_{13} \) in the diagram. The width of the measured luminescence spectrum, 10 meV, can be used to determine the various transition broadening terms [22]. At a field of 69 kV cm\(^{-1}\), the calculated broadenings are \( \hbar \tau^{-1}_{12} = 5.0 \text{ meV} \), \( \hbar \tau^{-1}_{23} = 2.2 \text{ meV} \), and \( \hbar \tau^{-1}_{13} = 8.5 \text{ meV} \).

3. Gain spectrum

The gain spectrum is derived from \( \frac{d\rho}{dE} = \frac{1}{2} [\rho, H] - S \), where \( \rho \) is the density matrix, \( H \) is the Hamiltonian matrix, and \( S \) is a phenomenological matrix introduced to characterize dephasing of each element of the density matrix [16–19, 27]. For the XTR model, these matrices can be written as

\[
\rho = \begin{pmatrix}
\rho_{11} & \rho_{12} & \rho_{13} \\
\rho_{21} & \rho_{22} & \rho_{23} \\
\rho_{31} & \rho_{32} & \rho_{33}
\end{pmatrix},
\]

Figure 2. Illustration of the energy levels and relevant parameters for calculating the gain spectrum using the XTR model.
and is a function of the photon density; when $D_i = 0.2$ and $0.1$ nm for the $3d$ transitions, coefficient is proportional to $\text{Im}(\rho_{12} e^{-i\omega t})$ and $\text{Im}(\rho_{13})$ as follows:

\[
\text{Im}(\rho_{12}) = -\frac{\Omega_t}{2} \frac{\eta_2 (\rho_{11} - \rho_{22})}{[(1 + \Omega_t^2 - \Delta(\Delta - \delta_{12}))\eta_1 \eta_3] + [\Delta \eta_2 + (\Delta - \delta_{12}) \eta_3^2]} \times \left\{ \frac{[1 + \Omega_t^2 \eta_2 \eta_3 + (\Delta - \delta_{12})^2 \eta_1^2] \eta_2 \eta_3}{(1 + \delta_{12}^2 \eta_3^2)(\eta_1 - \eta_2) + 2 \Omega_t^2 \eta_3(\eta_1 + \eta_3 - \eta_2)} \right\}.
\]

where $\tau_1, \tau_2, \tau_3$ are the total lifetimes of levels 1, 2, and 3, respectively.

The normalized gain spectra for both the INJ and XTR models at several electric fields are shown in Figure 3. At all fields, the calculated gain spectrum using the INJ model exhibits a single peak; this is in agreement with prior results using this model to study interface roughness broadening in QCL gain spectra [16]. The single peak is associated with the optical transition between u.l. and l.l. A second peak associated with the i.l. to l.l. transition is not present because of large dephasing due to interface roughness scattering and relatively weak coupling between the i.l. and u.l. The XTR model however indicates that the gain spectrum is more complicated than a single peak, which is in agreement with the original experimental results in [22]. For example, at 69 kV cm$^{-1}$, the spectrum exhibits a low-energy shoulder around 125 meV that is also observed in experiment. According to the density matrix model, emission at these lower energies is a combined result of strong coupling between the i.l. and e.l. and relatively small dephasing. Additionally, the coupling between the i.l. and e.l. results in a small spectral shift in the peak gain energy for the XTR model compared to the INJ model.

Figure 3. Calculated gain spectra under different applied electric fields for the INJ model (red, dashed) and the XTR model (blue, solid). An electroluminescence broadening of 10 meV at $\delta_{12} = 0$ is assumed. The gain is normalized so that the peak gain is unity.

We examine the influence of changes to the interface roughness on the gain spectrum and consider the limitations of the rate equation approach when evaluating spectra. Recent work has demonstrated a 15% difference in the width of electroluminescence spectra for mid-infrared QCLs operated under different polarity due to asymmetry in the interface roughness [28]. In this work, $\Lambda$ was 6 nm and $\Delta$ was 0.2 and 0.1 nm for the rough and smooth interfaces, respectively. We vary the interface roughness characteristics in equation (2) over a range similar to the difference between the rough and smooth interfaces and calculate the gain spectra using the XTR model. For each calculated gain spectrum with fixed $\Lambda$ and $\Delta$, the transition broadenings are all scaled by the same amount. Figure 4(a) depicts the gain spectrum versus $\eta_3$ and figure 4(b) shows three separate curves. Here, we use $\eta_3$ since it is most closely related to broadening of the low-energy peak and inversely proportional to the scaling factor for interface roughness. For longer $\eta_3$, the low-energy shoulder ($\sim 125$ meV) predicted by the XTR model increases in amplitude and narrows.

To demonstrate the limitations of the rate equation model, we numerically fit the gain spectra, $g(E)$, calculated by the XTR model using the method in [14]. Here, we use the sum of two Lorentzian lineshape functions that are weighted by the oscillator strength for the $1\rightarrow 2$ and $1\rightarrow 3$ transitions, $f_{12}$
and $f_{13}$, respectively:

$$ g(E) = f_{12} \frac{\gamma_{12}/\pi}{(E - \hbar \omega_{12})^2 + \gamma_{12}^2} + f_{13} \frac{\gamma_{13}/\pi}{(E - \hbar \omega_{13})^2 + \gamma_{13}^2}, $$

where $\gamma_{ij}$ is the FWHM of the individual transition. We use the oscillator strength, FWHM, and center energy of each Lorentzian as fitting parameters. The oscillator strength and FWHM of the secondary peak are shown in Figure 5 for different $\tau_{13}$. As expected, the FWHM scales inversely with $\tau_{13}$, and a 15% increase in $\Delta$ corresponds to a 11% decrease in the FWHM. Unexpectedly, the recovered oscillator strength also depends inversely on $\tau_{13}$; a 15% change of the scaling factor results in a 17% reduction of the fitted oscillator strength or an 8% change of the dipole. This later result shows that in the rate equation approach, the recovered oscillator strength of the diagonal optical transition depends on the interface roughness. This outcome is a result of coherent coupling between the l.l. and e.l., a process where the phase of the wavefunction must be considered, and is not predicted in a rate equation model which describes electron transitions as scattering events with an effective lifetime. Attempting to account for this outcome by modifying the effective lifetimes obscures the underlying physics, complicates analysis of the active region structure, and enables bad design practices. Indeed, prior to this work, this outcome was not a known limitation when using a rate equation analysis. Differences in design or growth conditions could result in devices with different interface roughness characteristics, and using the rate equation approach to interpret spectra could lead to confusion over subband populations and oscillator strengths. Furthermore, designing structures using the rate equation approach, overlooking the effects of coherence and dephasing, would face similar limitations. Such limitations could be particularly relevant for broad gain and nonlinear QCLs where many optical transitions are engineered between multiple subbands.

These results demonstrate the importance of scattering due to interface roughness on device properties of mid-infrared QCLs that depend on coherent phenomena, such as gain. This result is significant because the design of the heterostructure influences both the coupling and the scattering rate, and the effects of both on device properties have to be considered. For the active region examined in this paper, the gain spectrum can be considerably altered by changing the scattering times arising from interface roughness. In a real device, interface roughness is determined in part by growth quality; however, equation (2) also suggests another possibility for engineering the wavefunctions. Namely, interface roughness broadening between two states is influenced by the degree of spatial overlap, spatial extent of the
wavefunctions, and the number of interfaces crossed, and careful
design may reduce some of the effects of interface roughness.

4. Conclusion

In this paper, we have developed a three-level density matrix
model that describes the gain shape of a QCL active region that
incorporates multiple transitions. The results presented
here demonstrate the importance of considering coherence and
dephasing when designing active regions that incorporate
multiple transitions. Typical design strategies that use sta-
tionary states with rate equations may not represent actual
device performance because dephasing and coherence can
significantly alter the optical and electrical properties. These
results could have significant impact on broadband QCLs and
nonlinear QCLs where nearly-resonant wavefunctions are
routinely used to broaden the gain spectrum or engineer a
large nonlinear susceptibility.

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