Theoretical analysis of spectral gain in a THz quantum cascade laser: prospects for gain at 1 THz

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Abstract

In a recent Letter [Appl. Phys. Lett. 82, 1015 (2003)], Williams et al. reported the development of a terahertz quantum cascade laser operating at 3.4 THz or 14.2 meV. We have calculated and analyzed the gain spectra of the quantum cascade structure described in their work, and in addition to gain at the reported lasing energy of ≃ 14 meV, we have discovered substantial gain at a much lower energy of around 5 meV or just over 1 THz. This suggests an avenue for the development of a terahertz laser at this lower energy, or of a two-color terahertz laser.

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Following on the initial successful development of quantum cascade lasers in the mid-infrared, the field has continued its advance with the recent appearance of terahertz (far-infrared) quantum cascade lasers. The initial examples of these terahertz lasers, operating at \( \approx 4.5 \) THz, were based on chirped superlattice structures. An alternative design, operating at 3.4 THz, and based on a simple four well (per period) structure was recently reported by Williams et al. The simplicity of this structure provides a testbed for a nonequilibrium Green’s function theory we have recently developed to determine the nonequilibrium stationary state of quantum cascade laser structures operating under an applied voltage. This theory enables us to analyze transport and gain properties of these structures, giving us a tool to evaluate current-voltage (I-V) characteristics and nonequilibrium distribution functions, and to estimate level populations and lifetimes. In addition, we can use these parameters as a basis for evaluating and analyzing gain and absorption spectra in these structures. We have applied this theory to the structure reported by Williams et al., and we report here the results of our theoretical analysis. We highlight especially the new finding of substantial gain at the low energy of \( \approx 5 \) meV, well below the spectral region considered in Ref. 3.

Figure 1 shows the conduction band profile of two periods of the structure reported in Ref. 3 with an applied bias of 64 mV/period or 12.2 kV/cm. The Wannier-Stark wavefunctions (modulus-squared) are also shown. The energetic positions of the wavefunctions shown in the figure are the Wannier-Stark energies renormalized by the mean-field due to electron-electron scattering. Following Ref. 3, we designate levels 4 and 5 as the lower and upper laser levels respectively. In addition, we label level 1 (and 1’ for the neighboring period) the lower collector level, and level 2 (and 2’) the upper collector level.

We calculated the gain spectra by considering a simple two-level model interacting with a classical time-dependent electric field which gives the gain coefficient

\[
g(\omega) = -\frac{\omega \pi}{cn_B \epsilon_0 L_p} \sum_{(E_j > E_i)} |d_{ij}|^2 (n_i - n_j) L_{ij}(\omega),
\]

where we sum over contributions from each pair of levels \( i \) and \( j \). \( L_p = 52.4 \) nm is the length of one period of the structure, and \( n_B = \sqrt{13} \) is the background refractive index. The Lorentzian \( L_{ij}(\omega) = (\Gamma_{ij}/2\pi)/[(\hbar \omega - \Delta E_{ji})^2 + (\Gamma_{ij}/2)^2] \), with \( \Gamma_{ij} = \Gamma_i + \Gamma_j \), and \( \Delta E_{ji} = E_j - E_i \). The populations, e.g., \( n_i \), and the broadening parameters, e.g., \( \Gamma_i \), for level \( i \) are extracted from the nonequilibrium Green’s functions as described in Ref. 4. The populations and broadening parameters, as well as the energy differences \( \Delta E_{ji} \) and dipole matrix elements \( d_{ij} = e \xi_{ij} \) relevant for the following discussion are given in Table I. In addition to the shift due to the mean-field potential, the energy differences given in Table I include also the renormalization due to electron-phonon and interface roughness scattering. We neglect impurity scattering since the doping density per period, \( n_e = 2.8 \times 10^{10} \) cm\(^{-2} \), for this structure is low.

Figure 2 shows the calculated gain spectra at 30 K for three applied voltages, 64, 66, and 68 mV/period. At each of the two lower voltages, two strong gain features are seen: one lying between 10 and 15 meV, the other between 5 and 8 meV. There is a blue shift of the gain features as the bias increases in agreement with the experimental data. At 68 mV/period, the two gain features merge giving
a single broad gain feature stretching from \( \simeq 5 \) to 17 meV. To see the origin of these features, we look at the contributions to the spectra from transitions between each pair of levels.

Figure 3(a) shows the main transitions contributing to the gain for the applied voltage 64 mV/period. Contrary to the assignment given in Ref. 3, we find that the strong gain feature at around 12.6 meV is due to the transition between the lower collector level (1’) and the lower laser level (4), and not between the upper and lower laser levels. In fact, at this bias (see table I), the population in the lower laser level (4) is still slightly larger than the population in the upper laser level (5), giving rise to absorption rather than gain between these two levels. The dipole matrix element between levels 1’ and 4 (|\( z_{1'4} \)| = 3.2 nm) is of the same order, if slightly smaller than that between levels 4 and 5 (|\( z_{54} \)| = 5.14 nm). As importantly, however, there is a large population in the lower collector level 1’ (in fact, the most highly occupied level) and this gives rise to a substantial population inversion between levels 1’ and 4.

The strong gain feature at around 5.5 meV arises from the transition between the upper collector level (2’) and the upper laser level (5). This transition is favored by the large dipole matrix element (|\( z_{2'5} \)| = 6 nm), larger than that between levels 4 and 5. The large population in 2’ again gives rise to a significant population inversion.

Figure 3(b) shows the gain contributions at 66 mV/period. As in the previous case, the main contributions to the gain arise again from the transitions between levels 1’ and 4 (13.6 meV), and 2’ and 5 (6 meV). In addition, there are two smaller gain peaks. One peak at 15.2 meV can be assigned to the transition between levels 4 and 5, i.e., the lasing transition considered in Ref. 3. The other small peak at 7.8 meV is assigned to the transition between the collector levels 1’ and 2’. At the highest bias (68 mV/period) in Fig. 2, the contributions from these two latter transitions (4 – 5 and 1’ – 2’) become stronger leading to the double peaked structure seen at \( \simeq 15 \) meV, and the shoulder at \( \simeq 8 \) meV. The blue shift of the overall gain features with increasing bias can therefore be attributed to the appearance of these secondary peaks, as well as to the Stark shift mentioned in Ref. 3.

In addition to the gain features discussed above, the calculations also show a strong gain feature at around 2 meV at the lower voltage of 48 mV/period. This feature disappeared as the bias was increased. However, its reappearance is seen again in the low energy region of the 68 mV/period spectrum in Fig. 2. At a higher voltage of 70 mV/period this feature (originating from the 1’ – 5 transition) becomes stronger.

Thus, a theoretical analysis of this structure shows a complex behavior of the gain spectra, and their dependence on the applied voltage. Several transitions contribute to the gain spectra, with the spectral position and strength of each contribution depending sensitively on the applied bias. This suggests the possibility of generating gain in a spectral region stretching from \( \simeq 2 – 17 \) meV, with, in addition, some ability to tune the wavelengths at which the gain is enhanced by varying the applied bias.

We discuss next the robustness of our results to changes in the parameters used in the calculation. The values we have used for the broadening parameters \( \Gamma \) tend to overestimate the intersubband relaxation rate because \( \Gamma \) also includes the effect of intrasubband scattering processes. We have, however, also used an alternative approach to calculate the gain spectra, in which the linear response of the Green’s
functions and self-energies to an optical field are evaluated directly. This approach, which does not use the Lorentzian lineshape function, and hence does not require the parameters \( \Gamma_i \), reproduces the same gain features, in particular, the strong peaks around 6 meV, and between 10 – 15 meV. We have also repeated the calculations at 5 K, and including one extra level (i.e., considering six levels instead of five), and the main results of the above analysis are reproduced. The band structure parameters we used to describe the superlattice potential [conduction band offset (66%): \( \Delta E_c = 0.15 \text{ eV} \); electron effective masses: \( m_e^w = 0.067 \) in well, \( m_e^b = 0.07945 \) in barrier] are extracted from Ref. 7. The resulting theoretical current densities are approximately double the experimental values reported in Ref. 3. If, however, we use an 80% conduction band offset (\( \Delta E_c = 0.1825 \text{ eV} \)) as reported in Ref. 3, the calculated current densities are reduced, coming in close agreement with the experimental measurements. The main gain features discussed above are still present but slightly red-shifted, e.g., for 0.064 mV/period, the 5.5 meV feature in Fig. 3(a) is shifted down to 4 meV (\( \approx 1 \text{ THz} \)). The height of this gain feature increases by \( \approx 50\% \), while the height of the higher frequency gain feature decreases by \( \approx 25\% \).

We have reported here a detailed analysis of the gain spectra of a THz quantum cascade structure described in a recent Letter. The gain spectra exhibits a complicated behavior and dependence on the applied voltage, with several different transitions contributing to the gain. Surprisingly, besides the main lasing transition designated in Ref. 3, there are strong contributions to the gain from neighboring transitions, particularly transitions between the collector and upper or lower laser levels. These contributions are in fact larger than that of the designed lasing transition. The origin of these additional contributions to the gain is a sufficiently large overlap of the wavefunctions in the collector levels with those of the upper or lower laser levels to give dipole matrix elements comparable in magnitude to that of the lasing transition, as well as the large accumulation of population in the collector levels giving rise to a substantial population inversion with respect to the upper or lower laser levels. A notable finding is the appearance of a strong gain feature at around 5 meV, suggesting that this cascade structure design could also form the basis for a laser operating at around 1.2 THz (\( \lambda \approx 250 \mu \text{m} \)). There was no consideration or mention of this long wavelength spectral region in Ref. 3, which focused on the lasing emission around 3.4 THz. Thus, the results we report here urge more experimental investigations on this interesting structure, both for the prospect of a THz laser at a longer wavelength than achieved to date for any other quantum cascade laser structure, and as a test and verification of our theoretical approach and its predictive power.

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TABLE I: Parameters used in gain calculation for 64 and 66 mV/period. $n_i/n_e$ is the fractional population (total population normalized to 1) in level $i$. $\Gamma_i$ is the broadening parameter (in energy units) for level $i$. $\Delta E_{ji}$ is the energy difference between levels $j$ and $i$. These parameters are extracted from the quantum kinetics theory. $d_{ij} = e|z_{ij}|$ is the dipole matrix element between wavefunctions of levels $i$ and $j$.

| Parameter | 64 (12.2) | 66 (12.6) |
|-----------|-----------|-----------|
| $n_1/n_e$ | 0.35      | 0.26      |
| $n_2/n_e$ | 0.33      | 0.36      |
| $n_3/n_e$ | 0.16      | 0.16      |
| $n_4/n_e$ | 0.08      | 0.096     |
| $n_5/n_e$ | 0.077     | 0.12      |
| $\hbar/\Gamma_1$ (ps) | 0.94 | 0.71 |
| $\hbar/\Gamma_2$ (ps) | 0.99 | 0.81 |
| $\hbar/\Gamma_3$ (ps) | 0.71 | 0.79 |
| $\hbar/\Gamma_4$ (ps) | 0.44 | 0.44 |
| $\hbar/\Gamma_5$ (ps) | 0.5 | 0.55 |
| $E_{54}$ (meV) | 14.2 | 15.1 |
| $E_{1'4}$ (meV) | 12.5 | 13.4 |
| $E_{2'5}$ (meV) | 5.5 | 6.0 |
| $E_{32}$ (meV) | 39.4 | 39.4 |
| $|z_{54}|$ (nm) | 5.14 | 4.6 |
| $|z_{1'4}|$ (nm) | 3.2 | 3.8 |
| $|z_{2'5}|$ (nm) | 6.1 | 5.9 |
FIG. 1: Conduction band profile of structure reported in Ref. 3 with an applied voltage of 12.2 kV/cm, and Wannier-Stark levels. Wavefunctions (modulus-squared) are also shown.

FIG. 2: Calculated gain spectra at 64, 66, and 68 mV/period.
FIG. 3: Main contributions to gain spectra. (a) 64 and (b) 66 mV/period.