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Robust Density Matrix Simulation of Terahertz Quantum Cascade Lasers

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Abstract—A common setback to electron transport models for quantum cascade laser active regions is the inability to freely simulate widely varying designs. One solution to this problem is to use a density matrix formalism with a generalized treatment of scattering, wherein the well-defined energy eigenbasis is used, and the relative simplicity of the density matrix can be taken advantage of for rapid simulations. We have developed such a model from first principles in the past, and now built a simulator for terahertz quantum cascade lasers that calculates a fully self-consistent solution to the coupled problem of bandstructure, lasing field strength, and space charge. This level of depth enables us to examine the model’s performance across much of the design space and operating temperatures, for which we find generally good agreement. Areas for future improvement of the model are discussed, particularly the treatment of electron-electron scattering and continuum leakage. The model also enables us to make qualitative insights into the microscopic workings of the active regions, such as the non-equilibrium subband distributions and their response to the optical field, and the possibility for using two sequential optical transitions.

Keywords—terahertz, quantum cascade, density matrix, electron transport, III-V quantum wells.

I. INTRODUCTION

THz quantum-cascade (QC) lasers are a leading practical source of radiation over approximately 1-5 THz which rely upon intraband radiative transitions between electronic subbands within III-V heterostructure quantum wells. Record output power has reached 2.4 W in pulsed mode (1% duty cycle) [1] and 230 mW in cw [2]. These were, however, at a heatsink temperature of 10 K; at 77 K, the records are 1.8 W for pulsed [1] but only 5 mW in cw [3], [4]. In addition, the temperature record for any lasing has remained at 199.5 K for over half a decade [5]. Therefore, there is still much room for improved performance in areas such as power, temperature, tunability, broadband operation, and dispersion control, as well as to expand the higher performances across the THz spectrum, for example to the longer wavelengths.

Simulation of electron transport and gain in the active region still holds an important place in THz QCL design, and a wide variety of theoretical tools have been developed over the years, all with their respective benefits and limitations. Beyond the simple analytic models, the existing simulation

tools include ensemble Monte Carlo (EMC), density matrix (DM), and nonequilibrium Green’s Function (NEGF) methods. The EMC simulations use a semiclassical hopping picture of electron motion, which is relatively light computationally and can readily handle electron-electron scattering, but at the same time neglects important effects such as coherent tunneling and optical response [6]. The NEGF simulations offer the most complex and complete description of transport, but their considerable computational burden makes rapid simulation of active regions difficult, especially in the presence of a strong optical field [7], [8]. Still, NEGF has been widely used to simulate and provide useful details on the inner workings of QCLs [9].

For rapid simulation, a good balance is struck by the DM methods, which explicitly take into account coherences to reproduce important tunneling effects at a relatively low level of complexity. In fact, the original theoretical conception of a QCL used a simple analytic, phenomenological density matrix model to describe tunneling [10], which was later extended to study various simple designs [11], and then further for intense optical field [12], [13]. This type of model was used to assist the design of the current temperature record holder, although an EMC method was found to better describe the gain spectra [5]. Descendants of these methods are generalized numerical simulators handling an arbitrary number of states; this has enabled the study of certain THz QCL designs [14], [15], as well as more exotic devices including quantum dot cascade lasers [16], THz difference frequency generation in mid-IR QCLs [17], and self-mixing in a THz QCL [18].

However, a setback to DM simulations of this type is the reliance on phenomenology to reproduce the effects of electron localization by scattering. This is commonly done by defining, *ad hoc*, a set of “tight-binding” states spatially localized on either side of an injection barrier. This can be convenient in specific scenarios, particularly in mid-IR active regions when a single injection barrier is clearly the bottleneck to electron transport. It is less convenient when such a single barrier cannot be identified, and one cannot reasonably choose a well-justified set of “tight-binding” states. Furthermore, such models use a fundamentally incomplete treatment of scattering in the first place (based on Fermi’s Golden Rule transitions), and in a versatile model, arbitrary basis choices should not have to be made at all. This problem has been addressed in only a few works, by the derivation, from first-principles, of a more generalized scattering superoperator which couples all density matrix elements to one another [19], [20], [21], [22], [23]. As a result, the model becomes more complex, but the

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benefits are clear: use of the better-defined energy eigenbasis removes the need for the scientist to guess, *a priori* and in every specific case, where to force localization into the model.

In this paper, we employ a newly developed DM model of transport and gain that calculates generalized scattering [23], and demonstrate its wide applicability across the THz QCL design space enabled by the unnecessary for a phenomenological choice of localized wavefunction basis. With the aim to simulate lasing, we first consider the isolated effect of an optical field, finding that ordinary intensity can significantly thermalize subbands, as well as shift charge in the growth direction. The latter effect in turn couples the bandstructure itself to the lasing field via space charge, and therefore a true transport model of lasing action should incorporate all of these effects self-consistently. We implement such a scheme thanks to the numerical lightness of our DM model,¹ and simulate three exemplar designs of different character: one is a resonant-phonon design known for high temperature operation, another is a hybrid bound-to-continuum/resonant-phonon design known for high power, and a third is a scattering-assisted injection design operating at low-frequency. Simulations are performed over a wide range of temperatures reaching beyond lasing shut-off, and compared to key experimentally-measured parameters: current at threshold vs. temperature, current at maximum lasing intensity vs. temperature, and maximum operating temperature. To the best of our knowledge, this is the most comprehensive comparison of any DM-based model with experiment, and is made possible by our method's generality. We find mostly good agreement between theory and experiment, particularly for low temperatures and predictions of maximum operating temperatures. This thorough testing also enables us to identify limitations to the model, which we hypothesize lie mostly in the treatment of excited states and the continuum. Nonetheless, we argue that the results indicate the model's immediate usefulness for active region design. Furthermore, the model can assist the development of more novel devices such as frequency combs, owing to the simplicity by which coherent, nonperturbative optical response is calculated.

The paper is organized as follows: Section II provides an overview of the transport model, Section III gives an explicit example of the in- and out-of-plane redistribution of charge in response to a THz field, Section IV presents and discusses simulation results on three different active region designs, including comparison to experimental data, and Section V gives concluding remarks.

II. METHOD

A brief overview of the method is given here; for the full description, the reader is referred to [23]. Electrons are transported through the QCL active region by a complex interplay of tunneling, scattering, and interaction with an optical field. In general, we consider that the electron is influenced partly by (1) effects that we can write down a definite Hamiltonian H for, and (2) by other effects that are

only describable statistically. Effects under (1) comprise the “coherent” evolution, which is given in density matrix (ρ) context using the Liouville-von Neumann equation ($i\hbar\dot{\rho} = [H, \rho]$), while we resort to treating the “incoherent” effects under (2) perturbatively. The implied approximation is equivalent to the adiabatic approximation made in deriving Fermi's Golden Rule: specifically, that the incoherent effects act on the system over significantly longer timescales than the coherent effects, such that H sets the energy structure on top of which the incoherent effects act irreversibly.

The general equation of motion for the electron density matrix then has the following form:

$$\dot{\rho} = \frac{1}{i\hbar}[H, \rho] + \Gamma\rho, \quad (1)$$

where Γ is the generalized scattering superoperator which couples all density matrix elements to one another (i.e. $\dot{\rho}_{ab} = \sum_{cd} \Gamma_{ab,cd} \rho_{cd}$). The generalization of the scattering superoperator allows this formalism to capture important effects of scattering-induced localization that are not recovered by the phenomenological DM models which only include transitions and dephasing.

In the context of a QCL, or any quantum well intersubband device, the states are labeled with a subband index (i.e. m, n) and an in-plane wavevector (i.e. $\vec{k}, \vec{k}'$). Our formalism neglects coherences across different wavevectors (i.e. $\rho_{m\vec{k}, n\vec{k}'}$ for $\vec{k} \neq \vec{k}'$), amounting to an assumption of in-plane translational invariance. The remaining elements then hold in-plane wavevector resolution for all populations and coherences. Periodic boundary conditions are applied: all intraperiod density matrices are assumed identical, interperiod elements can exist and are themselves identical across different neighboring period pairs, and elements can affect one another across periods in ρ . ρ is allowed frequency content at dc and the frequency of an exciting optical field ω ; the complete steady-state is solved and the velocity is extracted at dc and ω to obtain current density and optical susceptibility. The direct steady-state solution of the equation of motion yields a coherent, nonperturbative account for the optical response, which enables study of a broad range of effects. In this case, the treatment is used to see the effects of strong optical field within the laser cavity, for both operating characteristics such as gain saturation and optically-driven current, and microscopic effects including the optically-induced in- and out-of-plane shifting of charge. We note that this nonperturbative treatment does in fact go to infinite order in the optical field (for example processes that follow $\chi^{(n)}(\omega = \omega - \omega + \omega - \omega + \omega - \dots)$ *ad infinitum*; however, it is not fully-inclusive in the sense that harmonics at $\pm m\omega$ for integers $m \geq 2$ are not accounted for. Discussion on similar topics can be found in [8], [17]; for the case of single-mode lasing considered here we do not expect such effects to be important.

For the full simulations of the active region performance in Section IV, iteration is necessary to account for space charge effects and an optical field clamped to an assumed threshold gain g_{th} . The transport solution holds the steady-state charge density, which is fed back to the Schrödinger-Poisson equation for a fully self-consistent transport calculation. Within

¹~10 min per fully self-consistent solution on a desktop (CPU: Intel Xeon E5-1650 v4 3.606 GHz)

each space-charge iteration, the optical field is also iterated to find the intensity under which the total gain at ω is $g(\omega) = g_{th}$, using a zero-finding (secant) method. The optical field is assumed to be homogeneous; therefore the simulation is effectively that of a plane wave traveling through the bulk active region medium, such that the computed performance metrics do not account for any spatial mode structure and are indicative of the active region alone.

The included scattering effects are of electrons with longitudinal-optical (LO-) phonons, ionized dopants, interface roughness, and alloy disorder. For the ionized dopants, Debye screening is used [24] with the electron temperature T_e approximated as 100 K greater than the LO-phonon temperature T_{LO} . The interface roughness height and correlation length are taken to be 2 Å and 10 nm, respectively. Notably, electron-electron scattering is missing in the present simulation tool due to its significant challenge being a many-body process affecting a single-particle density matrix, and discussion of its omission is included throughout the manuscript. Details on bandstructure calculation, including a method to include some excited states, are given in the Appendix.

III. OPTICAL REDISTRIBUTION OF CHARGE

It has long been known, and is immediately recognizable experimentally, that the lasing optical field has a significant impact on transport. This is manifested by a discontinuity in the differential conductance occurring at threshold, at which point stimulated emission suddenly becomes a contributor and enhances current flow through the device. Microscopically, the effect of the optical field on transport is more rich, as we will show that it both (a) powerfully thermalizes the subbands, as well as (b) significantly redistributes charge out-of-plane, which in turn changes the actual bandstructure through band bending.

Exemplary simulated results are shown in Fig. 1, for a device adapted from [25],[31] (bandstructure shown in Fig. 1a). This design is a hybrid resonant-phonon, bound-to-continuum design known for its high-efficiency and high-power operation. This design will be discussed in much more detail in Section IVa; here, we are focused on the effect of optical field on the in-plane electron distribution. For the purpose of demonstration, the effect of space charge is neglected, so that bandstructure is fixed and iteration is not performed. The gain spectra as the device is brought into injection resonance are shown in Fig. 1b; the gain peaks as well as absorptions from other transitions in the structure are visible.

Out-of-plane charge redistribution by the optical field is shown in Fig. 1c, comparing the electron densities obtained from steady-state solutions with no optical field (“Off”) and with a total optical intensity of 1 mW/ μm^2 (“On”). Even without the optical field, significant localization exists upstream of the tunneling injection barrier, despite the delocalized, near-perfectly-anticrossed appearance of the upper states (5, 6). This effect is held in the coherence between these two states, and arises naturally (i.e. with no *a priori* assumption of it) because of the generalized calculation of scattering. With the optical field included, the localization behind the barrier is

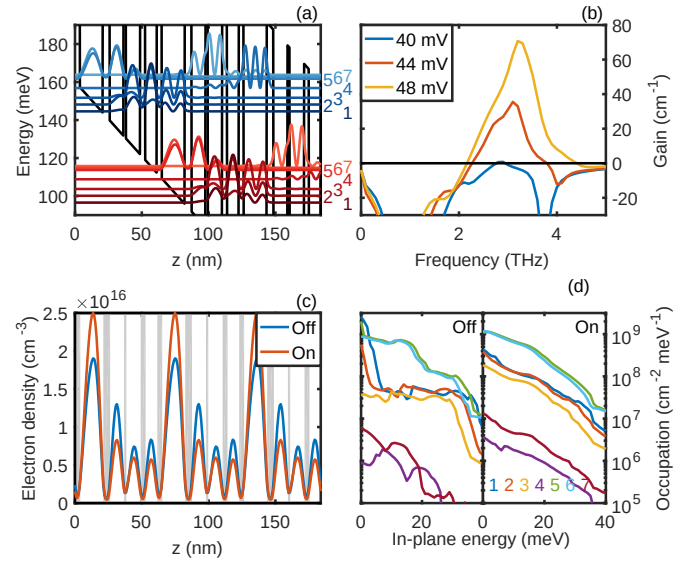


Fig. 1. Simulations at $T_{LO}=100$ K of the high-power device at ~ 3.3 THz demonstrated in [4], neglecting the effect of space charge on bandstructure. (a) Bandstructure at a bias of 48 mV, (b) Small-signal gain spectra at various biases leading to injection resonance, (c) steady-state electron densities obtained without (“Off”) and with (“On”) a strong optical field of 1 mW/ μm^2 at 3.3 THz, (d) Subband distributions for the same comparison.

further increased; this is the projection of gain saturation effects onto real space, as we see that less charge exists in the optically-active well. Furthermore, it appears that charge moves efficiently out of the lower state and further down the structure into the injector (widest) well, where it is eventually bottlenecked by the injection barrier, resulting in a higher steady-state charge density there. In more detailed simulations performed in the following sections, this redistribution of charge changes the potential itself, resulting in a changed bandstructure which is incorporated into the iteration algorithm.

Results are shown for the in-plane distributions of each subband in Fig. 1d, for the same comparison “Off” vs. “On”. In the Off-state, the distributions are markedly non-thermal (i.e. nonlinear on the log scale). In addition, it is clear by examining the slopes at the bottom of the subbands, where most of the population resides, that the upper states (5, 6) are far hotter than two of the lower states (1, 2). This aligns with the experimental results reported in [26], where upper states were estimated to be approximately 100 K hotter than lower states in a non-lasing scenario. It is quite interesting that this effect arises even without the inclusion of electron-electron scattering.

In the On-state, the subband distributions appear far more thermal, and additionally, seem to be much closer to each other in temperature (since the slopes are more similar); the extractor states are quite significantly heated to reach this condition. It seems that the optical field is driving the device towards a thermal equilibrium, all by loosening the bottleneck in transport that was intentionally designed into the optical transition. Furthermore, the populations in the excited states

TABLE I. SUBBAND ELECTRON TEMPERATURES (K) EXTRACTED FROM RESULTS IN FIG. 1.

Subband	1	2	3	4	5	6	7
Off	54	138	184	130	121	123	94
On	113	115	115	128	119	112	114

(4,7) are significantly larger with the optical field turned on, likely due to reabsorption of photons by electrons in the lower states. There is also a noticeable decrease in the occupation of all subbands at an in-plane energy of approximately the LO-phonon energy E_{LO} (36 meV in GaAs), although this is less significant in the presence of the optical field.

Nonthermal subband distributions in THz QCLs without an optical field have been predicted before, notably by density matrix simulations of [21], [22]. The thermalizing effect of the optical field was predicted by EMC simulations on a mid-IR QCL [27]; here, we have demonstrated the effect in a THz QCL where coherences (and thus at least a DM-based transport model) are important. These results can assist active region design for different functionalities: for example, in high temperature operation where the field is weak or non-existent, subband distributions can be expected to be highly nonthermal and at different temperatures, whereas in high power operation, the subbands can be expected to be well thermalized, including between each other. In the presence of weak or non-existent field, however, further investigation should focus on the possibility of subband thermalization due to electron-electron scattering, which is neglected in the present treatment.

Table I displays the subband electron temperatures extracted from the results in Fig. 1, $\langle E \rangle / k_B$ averaged using the subband occupations to weight. The thermalizing effect of the optical field is apparent; in the On-state, the temperatures of different subbands come closer together, settling mostly between ~ 15 -20 K hotter than T_{LO} . The subband temperatures are less well-defined in the Off-state where the distributions differ significantly from equilibrium; for example, the extracted temperature of subband 2 is quite hot even though it appears quite cold near its bottom by inspection of 1.

IV. SIMULATION OF VARIOUS DESIGNS

We now turn to a demonstration of the model's capability to simulate widely varying designs, using fully self-consistent solutions of bandstructure, transport, and optical field strength, providing good predictions of the actual performance. This is shown by a side-by-side comparison of three different designs in Figure 2: (A) a hybrid bound-to-continuum/resonant-phonon design that has been demonstrated to have very high power [4], similar to [25], (B) a resonant-phonon design for high temperature which holds the current T_{max} record [5], and (C) a scattering-assisted design for low frequency operation [28].

For the three designs, we performed simulations and extracted the light-current-voltage (L-I-V) characteristics at T_{LO} ranging from 100 to 300 K, as well as small-signal gain and subband distributions. For the L-I-V characteristics, the fully-iterated code for space charge and optical field was employed, assuming a threshold gain of $g_{th} = 20 \text{ cm}^{-1}$. The frequency

of the optical field was chosen specifically for each design, as indicated. Bandstructures are shown for the operating conditions indicated by the dots on the L-I-V characteristics, all of which were selected at $T_{LO}=100 \text{ K}$. For the small-signal gain, the bandstructures and scattering superoperators are taken at the result of the fully-iterated solution, for comparison's sake. The subband distributions are shown also for the operating condition marked by the dots: "On" refers to the fully-iterated solution, whereas "Off" indicates use of the same resultant bandstructure and scattering superoperator but without optical field. In all simulations, the in-plane energy resolution was set to 1 meV with a 70 meV limit. A line having the slope of a Boltzmann distribution at 100 K (T_{LO}) is added for comparison. Given the parameters outlined so far, simulation of different designs is quite simple from the viewpoint of the scientist using the code: only the different layer thicknesses, doping, and optical frequency need to be input, followed by identification of the basis states as shown in the Appendix.

For all comparisons with experimental measurements, some relevant differences must be kept in mind between the effective simulated scenario and the experimental one, beyond microscopic considerations such as exclusion of electron-electron scattering, leakage in the continuum, etc. These differences include:

- Assumption of uniform and monochromatic plane wave excitation: in the operating QCL, the optical field is highly non-uniform due to standing wave patterns, and can often be multimode as well. In addition, the actual lasing spectra may change significantly with bias in a Fabry-Perot cavity, although distributed feedback or VECSEL cavities can suppress such hopping.
- Assumption of a constant g_{th} : in reality, the actual loss is intricately dependent on the waveguide used, and the mode shape complicates the matter further. The loss also tends to vary with temperature [29].
- The temperatures in the simulation are the input LO-phonon temperature T_{LO} : especially in a resonant-phonon device, which is designed to emit LO-phonons for depopulation, T_{LO} is likely to be significantly hotter than the acoustic phonons to which they decay, which are in turn hotter than the heatsink. Furthermore, LO-phonon distributions may be significantly nonequilibrium in a real device [30]. No thermal feedback is included in the simulation either: effectively, this means that the laser is not heating itself as more electrical power is dissipated (i.e. temperature dependence on current).

In light of the thermal considerations, we choose to compare the simulated results only to pulsed measurements taken in metal-metal waveguides, the optimal conditions for heat extraction and low threshold. Regardless, the LO-phonons can certainly be expected to be hotter than the heatsink, especially at low temperature. To enable discussion, we will compare simulated currents at $T_{LO} = 100 \text{ K}$ against experimental currents at the lowest heatsink temperatures measured, and simulated currents at maximum simulated operating T_{LO} against experimental currents at maximum experimental heatsink temperature. Because of this and the other considerations, perfect

agreement is not expected given the unquantifiable differences between simulation and experiment, although reasonable agreement is certainly an important validation. Instead, the simulation should be viewed as a cheap test of the active region under simplified but physically meaningful conditions.

A. High-power design

This design strategy employs a tunneling injection occurring within the anticrossed doublet of states (6,7), followed by the radiative transition to the three coupled states below (1,2,4), which also perform tunnel extraction followed by LO-phonon emission to the injector of the next module. Two parasitic states (3,5) were also captured in the simulation. This design (and similar variants) is notable [31], as it holds records for highest peak pulsed power [1], [25] and cw power at 77 K [4].

We compare this device to experimental data reported in [4]. In a metal-metal waveguide configuration, the threshold current was 285 A/cm² at 7 K heatsink temperature, and the device lased up to 170 K, at which point the threshold current was 770 A/cm². In simulation, the threshold current at 100 K LO-phonon temperature of 296 A/cm² is quite close compared to the experimental measurement. Lasing was predicted up to $T_{LO} = 223$ K (interpolated). The threshold simulated current at maximum temperature is 759 A/cm², also quite close to the experimental result. Such close experimental agreement is likely by chance, however; for example, the J_{max} at 100 K was calculated to be 987 A/cm² in comparison to the experimental value of 730 A/cm². The overestimate of J_{max} is likely linked to an overestimate of optical intensity, which could be caused by either a gain overestimate or a loss underestimate. (The value of J_{max} is sensitive to the optical intensity, which decreases the upper state lifetime due to stimulated emission.)

Increase in current below threshold was predicted for temperatures up to 200 K, after which the current decreases; since this kind of behavior is not normally observed, this indicates that other mechanisms such as leakage to the continuum may become relevant above this temperature. Furthermore, at the lowest temperatures, a parasitic resonance is observed at ~ 36 mV bias which is not as prominent in the experimental data; it is not clear what shortcoming of the model allows this to emerge. NEGF simulations of a similar design, which included a model for electron-electron scattering, also predicted this feature to be more visible than was experimentally found [32].

Qualitatively speaking, the familiar linearity of the L-I characteristic is well-reproduced in the model. Interestingly, this is maintained despite the significant changes in the gain profile with bias - some blueshifting is observed, as is typically expected. The gain profile is also noticeably quite broad, tending to cover more than 1 THz. As this device is designed for 3.3 THz when biased to injection resonance, the gain peak is somewhat below this target for most of the dynamic range.

We note that an example of the effects of space charge is seen by comparison of these results with those shown in Figure 1 (where this same design was simulated without accounting for space charge). The differences are quantitatively significant: for example, the bias of injection resonance shifted

from approximately 48 mV to 52 mV with the inclusion of space charge, and the gain curves at these respective biases are quite noticeably different in both shape and peak height (peak gain is somewhat less with space charge than without, in this example). The situation of the extractor states in energy is also notably different. It is thus quite convenient, for the sake of design, that our model has a built-in capability to quantitatively account for the complex variety of effects related to space charge.

Effects of thermalization by the optical field are visible, although somewhat less dramatic than in Fig. 1 due to the smaller intensity obtained from the iterated solution. Additionally, now in this simulation including space charge, only state 1 is observed to be significantly cooler than the rest, as opposed to both states 1 and 2 in Fig. 1. It seems that subbands may reach this cold condition only when significantly populated. The excited state populations do increase significantly with the optical field, but are still quite small, at least at this temperature.

B. High-temperature design

This is a resonant-phonon design with a single-well injector. Tunneling injection takes place through the anticrossed doublet (4,5), after which the radiative transition is to two lower states (1,2) which are intended to anticross at approximately the same bias. Resonant-phonon emission is then used for depopulation. The two parasitic states we account for are visible at this bias as (3,6).

This design holds the current temperature record of 199.5 K (heatsink), at which point the threshold current was measured to be approximately 1.8 kA/cm²; at 8 K heatsink the threshold was measured to be about 980 A/cm². At low temperature ($T_{LO} = 100$ K), our threshold prediction is close at 827 A/cm². However, the discrepancy becomes worse with temperature, as we predict $J_{th} = 1.05$ kA/cm² at a maximum operating temperature $T_{LO} = 262$ K. Still, the prediction of maximum operating temperature itself appears quite reasonable: its status as a high-temperature performer is recognized by the simulation. At 100 K, J_{max} is predicted to be 1.48 kA/cm², reasonably close to the experimentally measured value of 1.61 kA/cm² at 8 K heatsink.

Immediately prior to lasing, a negative curvature $\partial^2 J / \partial V^2$ is visible in the simulation, as was seen in experiment. However, as was the case for the high-power design, this device also exhibits a subthreshold parasitic resonance at around 36 mV which was not visible in experiment. In addition, the subthreshold currents again generally do not increase with temperature beyond ~ 200 K, which again suggests an important influence of the continuum in drawing leakage current at high temperature.

The simulated L-I characteristics for this design are less linear than the high-power design, which we can attribute to the more complicated shifts in gain profile with bias. Indeed, in experiment, this device was found to lase between 2.6 and 3.22 THz, depending on both temperature and bias, and L-I curves deviating substantially from linear were measured. The extremely broad gain and complex bias dependence in

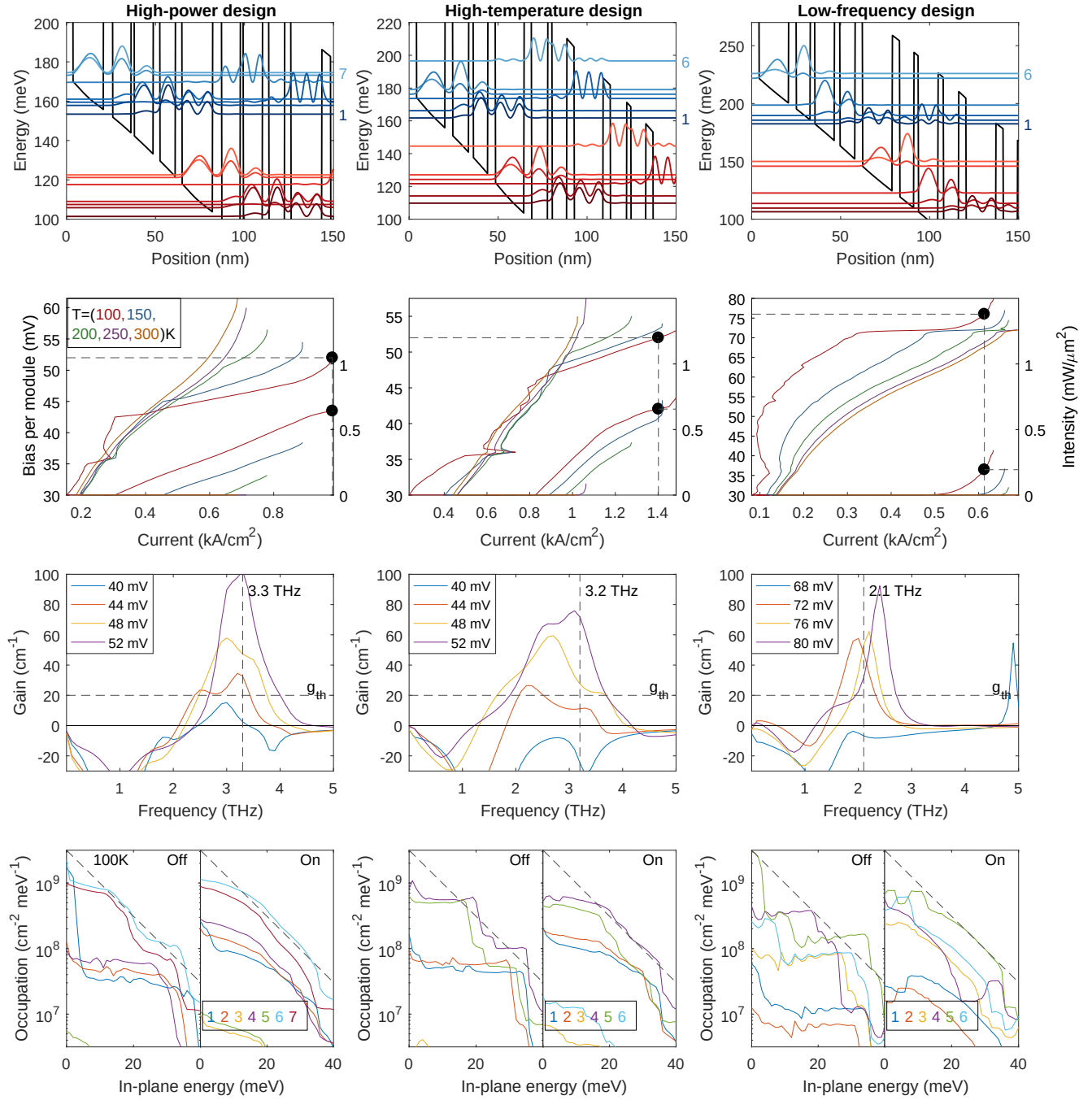


Fig. 2. Simulation results for three different THz QCL active region designs which demonstrated high-power [25], high-temperature [5], and low-frequency [28] operation. The operating conditions represented in the bandstructure and subband distributions are marked by the dots are for the three designs, at biases per module of 52 mV, 52 mV, and 76 mV. The frequencies marked on the gain profiles are those used for the full L-I-V simulations. In-plane energies were included in the simulation up to 70 meV, although subband distributions are shown only up to 40 meV. Gain spectra and subband populations are displayed from simulations at $T_{LO} = 100 \text{ K}$ in all cases. The subband distributions include a line having the slope of T_{LO} , for comparison.

this device can be attributed to the strongly tunnel-coupled and near-anticrossed nature of both the upper and lower laser level doublets. The strong tunnel-coupling is also to blame for

the high currents. This design was predicted to produce more power than the high-power design, for fixed T_{LO} ; however, it must be noted that it can be expected to heat itself more

significantly due to the considerably higher current levels.

Significant thermalization of the subbands is again predicted due to the optical field, and parasitic state populations are again notably quite small. Anomalously cool subbands were not predicted in this design.

C. Low-frequency scattering-assisted design

A significant improvement for low-frequency designs (less than ~ 2.5 THz) has been the so-called “scattering-assisted” design scheme, first demonstrated in [33]. In this scheme, electrons are injected into the upper state via LO-phonon scattering, rather than by resonant-tunneling injection. This avoids the difficulty of very large sub-threshold parasitic tunneling currents that occur when tunnel injection is used, as the radiative states become closer in energy. However, it comes at a cost of increased voltage drop per module, as well as the potential for reduced selectivity of injection (since LO-phonon scattering is less energy selective than tunneling). In the example of our model system from [28], tunneling occurs amongst states (5,6), after which the LO-phonon scattering injection occurs to (4). The intended radiative transition is then to state (3), which is close to tunnel-coupled with states (1,2). The spatially downstream part of this doublet has not ordinarily been pointed out in the literature; here, we will include its effect in the simulation. These states are then depopulated by resonant-phonon emission, such that two intended phonon emissions occur per module. We will make the argument that two optical transitions may in fact sometimes occur per module as well; the second being visible at this bias as from state (3) to the (2,1) doublet.

Predictions of current are significantly lower for this design; whereas we calculated a threshold of 492 A/cm^2 at $T_{LO}=100 \text{ K}$, the threshold current at 46 K heatsink was measured to be 750 A/cm^2 . At highest operating temperature of 144 K , the threshold current density was measured to be 1.2 kA/cm^2 , whereas our predictions were interpolated to only 675 A/cm^2 at maximum $T_{LO}=216 \text{ K}$. The observed difference between measured maximum heatsink temperature and simulated maximum T_{LO} is reasonable, and in-line with the other simulated devices. The model also successfully reproduced the subthreshold current plateau, visible in the IV curve at $T_{LO}=100 \text{ K}$ just above 100 A/cm^2 , which was observed in the experimental data at 46 K heatsink. In addition, the qualitative observation of little to no differential conductance discontinuity at threshold was reproduced in the simulation. This seems to be due to the already-high subthreshold differential conductance; the apparent discontinuity just before lasing is not related to lasing (this was verified by performing simulations that do not include an optical field).

Interestingly, this device seems to exhibit significantly larger dynamic range in experiment than in the simulation. A notable discrepancy is that the experimental data show the peak intensity occurring in a region of positive differential resistance, whereas in the simulation negative differential resistance occurs significantly before the alignment of the injector states (5,6). This suggests that the simulation is not capturing a transport mechanism which contributes excess conductance in

experiment and boosts the dynamic range. This is likely to be the continuum leakage, which we expect to be more severe in this active region, due to the notably higher bias with the same barrier heights as the other two; this would also explain the significant underestimation of currents in general.

At the subthreshold bias of 68 mV , a small gain peak is observed just below 5 THz , which is due to the laser acting in a way similar to the high-power design, but with higher frequency and worse injection alignment. As the bias increases, this peak shifts out beyond 5 THz , and the gain emerges at lower frequency. Some blueshift in this gain peak occurs with bias, which is consistent with the experimental data, in which lasing from 2.07 to 2.2 THz was observed. At the highest shown bias of 80 mV , yet another lower-frequency gain peak begins to emerge below 2 THz , which we can assign to inversion of the transition between state (3) and states (1,2). This is confirmed in the subband distributions. Subband distributions are significantly more nonthermal in this design than in the high-power and high-temperature designs. This includes colder-appearing states such as 1 and 5, but also less smoothness in all distributions. However, as electron-electron scattering is known to thermalize subband distributions, our neglect of it may have a significant impact on the overall L-I-V characteristic.

D. Overall discussion

In general, our model produces results that are close to the experimental measurements, to a degree similar to another survey performed using NEGF [9]. In addition, the model successfully identified qualitative aspects of the devices’ performance, for example: the superior temperature performance of the temperature-record design, gain spectra of the three devices and dependence on applied bias, near-linearity of L-I characteristics, differential discontinuity of the IV characteristics at threshold (or lack thereof in the low-frequency design), and the cooler temperature of subbands in specific cases. The model seems to perform well in predictions of threshold current at low temperature and of the maximum operating temperatures, while somewhat underpredicting current at higher temperatures, to a degree depending on design. Such performance of the model suggests that continuum or excited-state leakage could be an important factor for certain designs at higher temperatures, since this is not entirely captured in our implementation. Overall, the scattering-assisted design seems to have been the most difficult to simulate; as was speculated following a similar underestimate of current in [9], it seems that this device may carry a significant amount of current in the continuum due to the high bias applied. We speculate that this actually assists in electrical stability, improving dynamic range.

A visual of the key simulated currents (J_{th} and J_{max} vs. temperature) alongside experimental measurements is displayed in Figure 3. Notably, while simulations of the high-power design seem to have reproduced the temperature dependence well, higher-temperature current increase in the high-temperature design is largely missed (even though the currents at low temperature are quite accurate). The significant relative

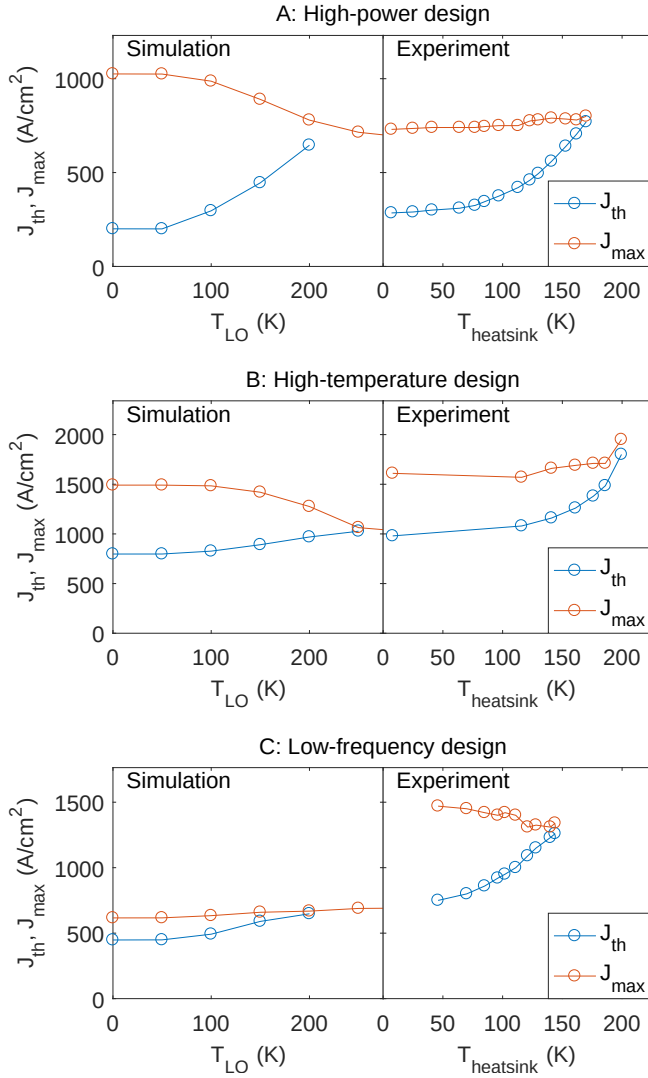


Fig. 3. Comparisons of simulated J_{th} and J_{max} vs. T_{LO} to their experimental counterparts vs. the heatsink temperature $T_{heatsink}$.

underestimate of current in the low-frequency design is clear, as well as the worsening with temperature, and the underestimate of dynamic range in the simulation. Table II displays the simulated maximum temperatures T_{LO} with the maximum experimentally-measured heatsink temperatures. Agreement is reasonable, as the maximum simulated operating T_{LO} values are only ~ 50 - 70 K hotter than the experimentally-measured heatsinks. The difference is smallest for the high-power device, which also draws the least current and would therefore be expected to heat itself the least.

V. CONCLUSION

A density matrix-based simulator of electron transport and gain was demonstrated for terahertz quantum cascade lasers that does not require the usual *ad hoc* choice of a localized basis. Instead, the method uses the better-defined energy

TABLE II. SIMULATED AND MEASURED MAXIMUM OPERATING TEMPERATURES.

Device	Maximum simulated operating T_{LO} (K)	Maximum measured operating $T_{heatsink}$ (K)
A: High-power	223	170
B: High-temperature	262	199.5
C: Low-frequency	216	144

eigenbasis, and approaches the treatment of scattering more generally such that all coherences scatter to one another, allowing scattering-induced localization to emerge on its own. Furthermore, the model is resolved in-plane, allowing the study of electron dynamics along those directions, and self-consistently solves for transport, bandstructure, and optical field all at once.

The model tends to reproduce experimentally measured currents with good accuracy; errors are roughly similar to that of more-established NEGF methods [9]. Some underestimates in threshold current, particularly at high temperature, may be due to transport mechanisms absent from the model, such as electron-electron scattering, or leakage to the continuum [35]. Still, the model was found to be a good predictor of the temperature performance, both relatively between designs and absolutely: the maximum LO-phonon temperatures in simulation tend to be between ~ 50 - 70 K hotter than the maximum heatsink temperatures measured. Qualitatively, the model reproduces most of the familiar features seen in the experimental L-I-V characteristics. L-I characteristics are usually near-linear, except in cases of dramatic gain shift with bias. The simulated gain spectra themselves also tend to agree nicely with the experimental measured spectra.

The model can also be used to study microscopies of operation, such as subband distributions. For all devices studied in this paper, we found that electron distributions can be highly nonequilibrium, and identified cases of anomalously cool subbands, as has also been observed experimentally [26]. The presence of an optical field seems to cause significant thermalization in all cases. However, further study in this area should include the effects of electron-electron scattering, which can be expected to thermalize the distributions further, quite possibly having significant effect on the macroscopic transport characteristics. The simulator can also be coupled with a thermal model to reproduce the effects of self-heating [34].

We conclude that our model is now ready for use as a general-purpose THz QCL active region simulator to aid design. By testing against a wide variety of experimental data, confidence has been established in the model's ability to identify qualitative trends that lead to enhanced performance such as higher temperature or power. Furthermore, its ability to see microscopically into the active region can help to provide inspiration for design directions: for example, the identified subband cooling effect might be used to boost temperature performance, or a low-frequency scattering-assisted design might be altered to fully exploit the benefits of two sequential optical transitions per module. Beyond conventional THz QCL operation, the model can also be expanded to handle optical

nonlinearities [17], for example to study four-wave mixing in QCL-based frequency combs [36]. The model could also be coupled with a model for cavity dynamics, and solved in the time domain, for similar problems [37].

APPENDIX A CHOICE OF HILBERT SPACE

Since our method is built upon use of the energy eigenbasis, we will now make clear precisely how this basis was identified. Although the QCL is a periodic structure, the actual states cannot be expressed as Bloch functions, because the existence of any bias field breaks translational invariance. Assuming independent minibands, each could be re-diagonalized in the presence of an electric field to form the so-called Wannier-Stark states which localize increasingly with the bias field; this description is useful in, for example, a superlattice at low bias [38]. However, in a QCL near design bias, coupling between the Wannier-Stark states from different minibands is crucial and central to the design strategies. Once this is included, the resulting true eigenstates become infinitely extended once again, as they mix not only with near-resonant other bound states, but with the continuum as well.

Due to effects of scattering-induced localization, this tends not to be a physical issue, but rather a numerical one. It is obvious by symmetry of the problem that an orthonormal set of eigenstates obeying periodicity in energy and position must exist; however, their full description requires infinite space. After the problem is inevitably bounded spatially to yield solutions of finite extent, the resulting solutions are orthogonal to one another but not necessarily to states in a period-translated set. This can create catastrophic problems in a method like ours which requires orthogonality between the basis states, unless care is taken to maintain a bounded and orthogonal set of states throughout a bias sweep. NEGF-based calculations do not encounter the same problem as energy eigenstates are not necessarily needed. In many NEGF QCL simulations, for instance, maximally localized Wannier functions of several adjacent device periods are used as the basis [38], [7]. However, energy-shifted periodic boundary conditions are typically used, which implies that only excited states confined within the simulated periods are fully included. Truly infinitely extended continuum states are difficult to fully capture in any method starting from a (semi)localized basis, whether the effective energy eigenstates used here or from a finite number of Wannier functions. The success of these methods might ultimately depend on whether the mean free path in the continuum is shorter than the simulated region. We also note an alternative method for studying QCLs using NEGF where only a single or few device periods are placed between dissipative contacts [39]; in this case, truly extended excited states may be recovered in the semi-infinite contact region, at the possible cost of missing some of the effects of periodicity.

Figure 4 illustrates the method we used to address this problem for the current work: essentially, we first identify a well-behaved set of states at a bias point near operation to serve as the solidified Hilbert space for one module, and then under

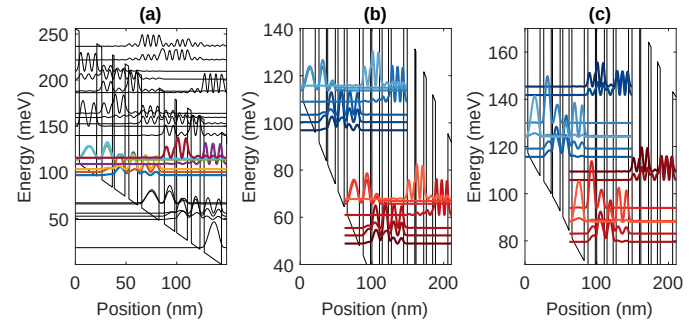


Fig. 4. Illustration of the process by which a Hilbert space of basis states is selected: (a) bounded solutions are found and a Hilbert space of subbands is identified, (b) these states are approximated to constitute the periodic set of solutions to the infinitely-extended problem, (c) under different potential due to bias or space charge, this Hilbert space is re-diagonalized assuming mixing only within the identified module (e.g. within blue states only).

different potentials, the solutions are re-diagonalized. Mixing is only allowed within the identified module so as to guarantee spatial restriction. There are two points of approximation: first, the choice of spatially-restricted states, and second, the neglect of mixing at different potential to other modules and with the continuum. This approach is well-suited to resonant-phonon designs, where it is sensible to neglect mixing across the large energy gap for depopulation. Choosing this gap as the energy boundary between modules also helps to keep intermodule non-orthogonality to a minimum.² Some number of higher-energy states (parasitic states which can carry current at high temperatures [35]) can be accounted for, depending on the spatial extent of the original bounded problem in (a); however, they become increasingly difficult to define as more states are included. Furthermore, nonparabolicity is neglected in the present formalism, which would be expected to affect the excited states more strongly. Nonparabolicity could be accounted for in-principle by diagonalizing the full three-band Kane model, so that orthogonality is retained [17], [40], [41]; such a treatment should also include the altered subband shapes and the scattering superoperator acting in all three bands.

While the requirement to choose a Hilbert space imposes somewhat of a limitation on the model, it is certainly less constricting than needing to choose an artificially localized basis, as in most previous density matrix models. Some of the limitations could be solved by a theory of generalized scattering that does not require the energy eigenbasis but is instead completely basis-invariant, so that well-localized basis functions can be chosen from the start as is done in NEGF methods. However, this would come with costs, such as significantly increased complexity and loss of the picture of actual energy eigenstates in the first place.

²A measure of this is $\sum_{ab} |\langle \psi_a^x | \psi_b^y \rangle|^2 / N$, where $\{\psi^{x,y}\}$ are the states in the two subspaces (modules in this case) x and y , and N is the number of states in each. This is equivalent to $\text{Tr}(P^x P^y)$, where $P^{x,y}$ are projection operators onto the two subspaces. Equivalent subspaces would have a value of 1 for this measure, whereas 0 would indicate complete orthogonality; for the bases selection shown in Fig. 4, we obtain a value of 6.5×10^{-4} .

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