Room Temperature Lasing of Terahertz Quantum Cascade Lasers Based on a Quantum Dot Superlattice

Alexander Mittelstädt, Ludwig A. Th. Greif, Stefan T. Jagsch, and Andrey Schliwa
Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany
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We investigate room temperature lasing of terahertz quantum cascade lasers using quantum dot chains as active material. Bandstructure calculations for such extended systems of coupled quantum dots are made possible by a novel ‘linear combination of atomic orbitals’–like approximation, based on single quantum dot wavefunctions. Our results demonstrate that terahertz quantum cascade lasers greatly benefit from intrinsic properties of quantum dots, such as reduced phonon coupling and in-plane scattering, enabling room temperature lasing with significantly reduced threshold current densities.

In conventional quantum cascade lasers (QCLs) electrons run down a staircase potential generated by a superlattice of coupled quantum wells. Amplification of radiation is realized via intra-band transitions of electrons, a concept first proposed by Kazarinov and Suris in 1971 [1]. Population inversion between the subbands is achieved by the meticulous engineering of electron lifetimes and transition probabilities by means of layer thicknesses and external bias, having the capability of tuning intra-band transitions with meV accuracy. Since its first realization in 1994 by Faist et al. [2], QCLs have become a well-established mid-infrared light source with applications in, for instance, trace gas analysis, medical diagnostics and optical communications [3, 4]. As biological tissue and many other materials, such as e.g. textiles, are transparent within the terahertz (THz) spectrum (far-infrared), QCLs are promising alternative light sources for non-invasive inspection and imaging [5, 6]. Since the first realization of a THz QCL in 2002, with operating temperatures of up to 50 K [7], constant development of device design and material growth has allowed operating temperature of up to 210 K, as recently demonstrated in Ref. [8]. However, operation at room temperature is still impeded by an increasing competition from non-radiative scattering losses and free carrier absorption [9–11], threshold current densities found in quantum-well based QCLs are generally high (∼kAcm⁻²), regardless of the operating wavelength.

To address these obstacles, QCLs with an active region composed of quantum dot (QD) chains have been proposed (QD-QCL) [15, 16], where the localized states in the QDs lead to reduced electron-phonon scattering (phonon bottleneck) and free-carrier absorption, increasing carrier lifetimes by orders of magnitude and thereby improving temperature stability [17–19]. QD-QCLs also benefit from the intrinsically narrow gain spectrum of QD-chains and significantly reduced threshold current densities are predicted, when compared to quantum well based QCLs [16, 20, 21]. So far, however, the literature on QD-QCLs is largely speculative and based on assumptions of the electronic structure of such extended systems of coupled QDs, while a rigorous simulation of the electronic structure of a realistic stack of QDs is still missing. In this letter, we present a bandstructure calculation for a stack of 20 QDs, featuring an intra-band staircase potential suitable for THz operation. Our QD-QCL design comprises a two-dimensional array of InGaAs QD-chains with a two-QD unit cell superlattice, embedded in a GaAs matrix, cf. Fig. 1. Strong electronic inter-dot coupling results in delocalized electronic states along the QD-chain, which are engineered to facilitate population inversion at a certain external bias. Subsequently, we estimate the threshold current density of such a QD-QCL at operating temperatures of 10 K and 300 K, using an empirical rate equation model. Based on realistic device parameters, we find a strongly reduced threshold current density, when compared to devices based on a quantum
well heterostructure, thus paving the way for the development of a THz QCL operating at room temperature. The InGaAs/GaAs material system is chosen as it is the most common in quantum well QCLs and provides both a mature growth platform and superior material quality \[5\] [10] [22].

The challenge of finding suitable parameters for the QD gain material boils down to the computational cost of simulating i) stacks of more than ten coupled QDs as well as ii) calculating dozens of excited electronic states of these QD-chains. In a parameter study, QD-chain length, tunnelling barrier width, and composition and geometry of the individual QDs are varied in order to realize the desired bandstructure. Although systems containing millions of atoms can be addressed via well-established empirical tight-binding and 8-band \( \mathbf{k} \cdot \mathbf{p} \)-models [23]–[27], most investigations involve just a stack of two QDs and a few involve nine layers of QDs calculating a maximum of twelve electronic states. Moreover, this expensive calculation would be necessary for every set of trial parameters for individual QDs and QD-chain. This makes simulations of stacks of ten and more QDs, including 50 and more electronic states impractical or even impossible. In order to drastically reduce the associated computational cost, we developed a novel ‘linear combination of atomic orbitals’-type of approximation, based on single QD single-particle wavefunctions, capable of calculating the electronic structure of such QD-chains. Our ‘linear combination of quantum dot orbitals’-method (LCQO) can be implemented on top of any atomistic or continuum model for the electronic states of the QDs and is in no way limited to the 8-band \( \mathbf{k} \cdot \mathbf{p} \)-wavefunctions exemplarily used herein. The LCQO method is introduced, along with a detailed study of the electronic properties of stacks of vertically coupled QDs, in (the associated manuscript) Ref. [PRB].

**METHOD OF CALCULATION**

The underlying idea of the LCQO method is to split a large system of coupled QDs, i.e. a large eigenvalue problem, into a set of subsystems of single QDs for which single-particle states can be efficiently calculated. Provided that \( \{ |\varphi_{n}^{I}\rangle \} \) denotes the eigenstates of subsystem \( I \), the set of basis functions of the large system \( M \) is found by a union of the eigenstates for all subsystems \( \{ |\varphi_{k}^{M}\rangle \} := \cup \{ |\varphi_{n}^{I}\rangle \} \). This composite basis is then used to expand the eigenstates of the LCQO system \( |\psi_{LCQO}^{M}\rangle = \sum_{k=1}^{m} a_{ik} |\varphi_{k}^{M}\rangle \), where \( m \) is the cardinality of \( M \). Adopting the Rayleigh-Ritz variational principle [28] for the energy functional results in a generalized eigenvalue problem

\[
\sum_{k=1}^{m} \left[ \langle \varphi_{l}^{M} | \mathbf{H} | \varphi_{k}^{M} \rangle - \varepsilon_{i} \langle \varphi_{l}^{M} | \varphi_{k}^{M} \rangle \right] a_{lk} = 0, \tag{1}
\]

yielding \( m \) eigenvalues \( \varepsilon_{i} \) and eigenvectors \( |a_{i}^{\pm}\rangle \), resulting in a set of LCQO single-particle functions \( |\psi_{i}^{LCQO}\rangle \).

Above, \( \mathbf{H} \) denotes the Hamiltonian of the composite system \( M \), where we consider the strain distribution and resulting piezoelectric fields in the whole heterostructure.

To make the LCQO efficient, first a library of single QDs varying in geometry and material composition is created, whose electronic single-particle states are subsequently calculated to serve as a basis in the LCQO calculations. The eigenvalue problem in Eq. (1) is then solved via a standard linear algebra package (LAPACK) routine.

The crucial benefit of this method is the reduction to a variational problem for finding the stationary points for \( \langle \mathbf{H} \rangle \) of the large system as a function of a finite set of coefficients, whereby the calculation time is reduced by at least three orders of magnitude, compared to a full 8-band \( \mathbf{k} \cdot \mathbf{p} \) calculation. A detailed derivation of the LCQO method, a performance test and a direct comparison to full 8-band \( \mathbf{k} \cdot \mathbf{p} \) simulations are provided in (the associated manuscript) Ref. [PRB].

The threshold current of the QD-QCL system is derived via an empirical three-level rate equation model, adapted from [21] [29],

\[
\frac{\partial N_{3}}{\partial t} = \frac{j(t)}{eL_{w}} + R_{2,3} - R_{3,2} - R_{3,inj} - R_{3,2}^{ph} \tag{2}
\]
\[
\frac{\partial N_{2}}{\partial t} = 3 \sum_{i=1,3} R_{i,2} + R_{3,2}^{ph} - R_{2,3} - R_{2,1} \tag{3}
\]
\[
\frac{\partial N_{1}}{\partial t} = R_{2,1} + R_{2,1}' - R_{1,2}' - R_{1,2}^{ph} - R_{1,2}' \tag{4}
\]
\[
\frac{\partial S}{\partial t} = R_{3,2}^{nd} + \beta R_{3,2}^{sp} - \kappa S, \tag{5}
\]

where \( R_{3,2}^{ph} \) is the sum of induced and spontaneous emission rates. The rate equations for the carrier densities \( N_{i} \) are connected to the occupation probabilities \( n_{i} \) via \( N_{i} = N_{i}^{*} n_{i} \), where \( N_{i}^{*} = 2\rho/L_{w} \), with the lateral density of QD-chains in the active region \( \rho \), the QD-chain length \( L_{w} \) and the number of cascades per QD-chain \( C \). The injection current density is \( j(t) \) and \( \epsilon \) is the elementary charge. The \( \beta \)-factor defines the fraction of spontaneous emission coupled into the laser mode and \( \kappa \) is the total cavity loss. The non-radiative in- and out-scattering rates are defined by \( R_{i,j} = N_{i}(1-n_{j})/\tau_{i,j} \), where the intradot relaxation times \( \tau_{i,j}(\Delta E) \) depend on the energy difference between discrete QD-states and are therefore considered constant. Carrier escape and relaxation times are related via a quasi-Fermi distribution at temperature \( T \)

\[
\tau_{i,j} = \tau_{i,j}^{0} \exp \left( \frac{E_{i} - E_{j}}{kT} \right). \tag{6}
\]

\( S \) is the photon density and the induced emission term is \( R_{3,2}^{nd} = S \gamma(n_{3} - n_{2}) \), with the gain coefficient \( \gamma \). The spontaneous emission term is \( R_{3,2}^{sp} = N_{3}(1 - n_{2})/\tau_{i,j}^{sp} \), where
Figure 2. Schematic illustration of two adjacent QD-chains of length $L_w$ separated by a distance $d$ greater than the exciton Bohr radius (electronically decoupled). The individual QDs in the chain are separated by barriers of width $b$.

the lifetime of the spontaneous emission is determined via Fermi’s golden rule $\tau^{-1}_{\text{sp}}(\epsilon) \propto \hbar^2 |\epsilon \langle \psi_j | \nabla | \psi_i \rangle|^2$. Here, $|\psi_i\rangle = |\psi_i^{\text{LCQO}}\rangle$ and $\omega_{ij}$ is the transition frequency. The rates marked by the dashes in Eq. 4 are related to the adjacent period’s QD-chain states, since the carrier density $N_1$ is equal to the density $N_3'$ in the upper laser level of the next period.

DISCUSSION AND RESULTS

The realization of a THz QD-QCL imposes specific requirements on the design of the bandstructure. In particular, these are (i) cascaded intra-band transitions of identical energy with electron probability densities shared by neighbouring QDs and (ii) in- and out-scattering rates that allow population inversion between the laser levels. Facilitated by the LCQO method and based on the results and discussions in (the associated manuscript) Ref. [PRB], we initially studied more than a hundred possible realizations of QD-chains at various external biases, with the prerequisite of using experimentally realistic parameters. The QDs are electronically coupled across tunnelling barriers of width $b$, forming QD-chains of length $L_w$, see Fig. 2. The individual QD-chains are electronically uncoupled through a lateral separation $d$ much larger than the exciton Bohr radius of the material system, i.e. $d \gg 15$ nm for GaAs [30]. The QD-chains consist of InGaAs QDs modelled as truncated pyramids, following TEM imaging of Stranski-Krastanov QDs [31, 32]. For the bandstructure presented in Fig. 3, we assumed In$_{0.7}$Ga$_{0.3}$As QDs with a side-wall inclination of 40°, a basis length of 14.7 nm and a height of 2.8 nm, in agreement with reports in [33, 34]. The vertical aspect ratio (height divided by base diameter) of $AR_v = 0.135$ is chosen slightly lower to account for material interdiffusion found in experiment. The resolution of the finite difference grid for the LCQO calculations is set to two monolayers (MLs) of GaAs (5.653 Å).

Instead of a uniform QD separation, we use a two-QD unit cell with QDs coupled through a $b = 8$ MLs barrier, with subsequent unit cells separated by $b = 20$ MLs, which leads to a more readily achievable population inversion for the characteristic transition. Fig. 3 shows a zoom into the bandstructure of a stack of 10 unit cells (20 QDs), which results in seven identical QCL periods along the QD-chain. As discussed in (the associated manuscript) Ref. [PRB], edge effects are essentially converged for chains of more than 10 QDs, after which the number of QCL periods increases with subsequent stacking. The laser transition is realized between the ground state $|3\rangle$ (s-type symmetry) of a two-QD unit cell and the neighbouring unit cell’s excited state $|2\rangle$ (p-type), as indicated by the red arrow in Fig. 3. The transition results in a photon energy of $\hbar \omega \approx 16$ meV ($\approx 3.8$ THz) at an external bias of $E_{\text{ext}} = 24$ kV cm$^{-1}$ with lifetime $\tau = 1.0$ ns at $T = 10$ K, as estimated by Zibik et al. for the characteristic p-to-s intra-dot carrier relaxation times of self-assembled InGaAs QDs [19]. The symmetries of the orbitals are as specified in Ref. [PRB]. The strongly coupled QDs enable a fast depletion of the lower laser level $|2\rangle$ ($\Delta E = 44$ meV) into the QDs ground state $|1\rangle$ (black arrow in Fig. 3). The energy spacing of 44 meV results in an efficient intra-dot relaxation process with $\tau_2 \approx 30$ ps at $T = 5$ K, as measured by Zibik et al. [18], due to a high LO-phonon density. Since this design facilitates a diagonal laser transition, its energy can be uniformly adjusted using the external bias. For example, by reduc-
Table I. Parameters used for the rate equation model

| parameter | physical meaning | value | Ref. |
|-----------|------------------|-------|------|
| $L_w$     | length of a QD-chain | 200 nm |      |
| $W$       | width of the cavity | 5.7 $\mu$m |      |
| $L$       | length of the cavity | 10.8 $\mu$m |      |
| $C$       | Number of cascades per chain | 7 |      |
| $m_e^*$   | eff. el. mass for In$_x$Ga$_{1-x}$As | 0.0356m$_e$ | [41] |
| $\kappa$  | total internal losses | 2.5e10 s$^{-1}$ | [42] |
| $g$       | gain coefficient | 9.6e10 s$^{-1}$ | [42] |
| $\beta$   | beta-factor | $10^{-3}$ |      |
| $n$       | mode index | 3.6 |      |
| $\tau_{3,2}$ | lifetime of $|3\rangle \to |2\rangle$ at $T = 10$ K | 1.0 ns | [19] |
|           | at $T = 300$ K | 60.0 ps | [42] |
| $\tau_{2,1}$ | lifetime of $|2\rangle \to |1\rangle$ at $T = 10$ K | 30.0 ps | [15] |
|           | at $T = 300$ K | 10.0 ps | [15] |

Figure 4. The upper and lower laser levels occupation probabilities $n_3$ (solid blue line), $n_2$ (dashed blue line) as a function of pump current density $j$ and the photon flux density (solid black line) at temperatures $T = 10$ K (a) and $T = 300$ K (b). $S$ is the photon flux density at a mode reflectivity of the facet of $R = 0.95$.

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In conclusion, we investigate room temperature lasing in a THz QCL with QD-chains as active material. Based on a novel LCQO approach, we are able to efficiently optimize the gain material and show a cascaded lasing transition within the THz spectral region, at $\sim 3.8$ THz. Our two-QD unit cell design allows a diagonal laser transition that is adjustable by external bias, while maintaining a fast depletion of the lower laser level independently. The crucial aspect to achieve room temperature operation is the strong suppression of non-radiative carrier losses and thermal backfilling by the quasi zero-dimensional nature of the QDs. A threshold current density at room temperature as low as $j_{th} \approx 3.0$ kAcm$^{-2}$ is predicted by a rate equation model based on our realistic bandstructure calculation. Our results serve as a valuable guideline for future experiments towards room temperature THz QCLs.

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