Are Quantum-Cascade Lasers really quantum?

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(March 22, 2022)

The first global simulation of semiconductor-based quantum-cascade lasers is presented; Our fully three-dimensional approach allows to study in a purely microscopic way —without resorting to phenomenological parameters—the current-voltage characteristics of state-of-the-art unipolar nanostructures. Based on the proposed theoretical scheme, we are able to give a definite answer to the long-standing controversial question: is charge transport in quantum-cascade lasers mainly coherent or incoherent? Our analysis clearly shows that a proper inclusion of carrier-phonon as well as carrier-carrier scattering within a semiclassical framework gives excellent agreement with experimental results.

72.10.-d, 72.20.Ht, 73.63.-b, 78.67.-n

Since the seminal paper of Esaki and Tsu \cite{1}, semiconductor-based nanometric heterostructures have been the subject of an impressive theoretical and experimental activity, due to their high potential impact in both fundamental and applied research \cite{2,3}. One of the main fields of research focuses on exploiting “band-gap engineering”, namely the splitting of the bulk conduction band into several subbands, to generate and detect electromagnetic radiation in the infrared spectral region, as already envisioned by Kazarinov and Suris \cite{4}.

Unipolar coherent-light sources like quantum-cascade lasers (QCLs) \cite{5}, are usually modelled in terms of three-level systems \cite{6}. Their theoretical description is thus often grounded on purely macroscopic models: the only relevant physical quantities are the various carrier concentrations within the different energy levels, whose time evolution is dictated by proper sets of coupled rate equations. This simplified three-level picture does not take into account the existence of transverse or “in-plane” degrees of freedom and thus provides no information on the microscopic processes governing carrier dynamics within the three-dimensional (3D) multi-subband structure. As pointed out in \cite{7}, such a macroscopic modeling can only operate as an a posteriori fitting procedure: The best-fit phenomenological parameters obtained through a comparison with experiments do strongly depend on details of the 3D hot-carrier distribution and therefore on the device operation conditions.

QCLs are complex devices, whose core is a multi-quantum-well (MQW) structure made up of repeated stages of active regions sandwiched between electron-injecting and collecting regions. When a proper bias is applied, an electron cascade along the subsequent quantized-level energy staircase takes place. For a detailed understanding of the basic physical processes involved, two main issues need to be addressed: (i) the nature of the hot-carrier relaxation within the device active region; (ii) the nature—coherent versus incoherent—of the physical processes governing charge transport through injector/active-region/collector interfaces. Point (i) has been recently addressed in \cite{7}, where the macroscopic treatment of hot-carrier relaxation within the device active region previously mentioned has been compared to a fully kinetic description, based on the following set of equations \cite{7}:

\[ \frac{d}{dt} f_{\mathbf{k}\nu} = [g_{\mathbf{k}\nu} - \Gamma_{\mathbf{k}\nu} f_{\mathbf{k}\nu}]_{i/l} + \sum_{\mathbf{k}'\nu'} \left[ P_{\mathbf{k}\nu,\mathbf{k}'\nu'} f_{\mathbf{k}'\nu'} - P_{\mathbf{k}'\nu',\mathbf{k}\nu} f_{\mathbf{k}\nu} \right]. \]

(1)

Here, the first two terms describe—still on a partially phenomenological level— injection/loss (i/l) of carriers with wavevector \( \mathbf{k} \) in subband \( \nu \), while the last ones describe intra- as well as inter-subband in- and out-scattering processes (\( \mathbf{k}\nu \rightarrow \mathbf{k}'\nu' \)) within the device active region only. The Boltzmann-like structure of (1) allows for a partially stochastic solution; to this aim, the commonly used Monte Carlo (MC) method \cite{8} has been adopted. The simulated experiments in \cite{7} show that, as far as the active region is concerned, the quantum-cascade process is mainly determined by carrier-optical phonon emission; Moreover, the values of effective inter-level rates strongly depend on the shape of the in-plane charge distributions as well as on Pauli-blocking effects.

However, the microscopic analysis in \cite{7}, being limited to the device active region only, does not allow to answer point (ii): which are the basic physical mechanisms governing charge transport through injector/active-region/collector interfaces? This issue is intimately related to the long-standing controversial question \cite{9}: is charge transport in quantum-cascade lasers mainly coherent or incoherent?

To provide a definite answer to this fundamental question, in this Letter we present the first global simulation—injector plus active region plus collector—of semiconductor-based QCL structures. To this end, the...
partially phenomenologic model in (1) has to be replaced by a fully microscopic description of the whole MQW core structure. This is not a trivial task. It requires: (i) a proper description of all the 3D electronic states within the active as well as injector/collector regions; (ii) a fully microscopic simulation scheme to “close the circuit”.

As a starting point, we evaluate —within a self-consistent Schrödinger-Poisson approach in an envelope-function framework (3) —the set of 3D single-particle electron states \( \{kv\} \) corresponding to a single QCL stage, e.g., active region plus collector/injector in the presence of the applied bias (see Fig. 1). Given such carrier states, we consider the ideal MQW structure obtained as infinite repetition of this QCL periodicity region (see Fig. 1). Within such extended scheme, the time evolution of the carrier distribution function \( f_{k\alpha} \) is governed by the following Boltzmann-like equation:

\[
\frac{d}{d\tau} f_{k\alpha} = \sum_{k',\alpha'} \sum_s \left[ P^{s}_{k\alpha,k'\alpha'} f_{k'\alpha'} - P^{s}_{k'\alpha',k\alpha} f_{k\alpha} \right].
\]

Here, \( \alpha \equiv (\lambda, \nu) \) denotes the generic electron state in our MQW structure, i.e., the \( \nu \)-th state of the \( \lambda \)-th stage, while \( s \) labels the different scattering mechanisms considered, e.g., carrier-phonon, carrier-carrier, etc.

To “close the circuit”, we impose periodic boundary conditions limiting the inter-stage (\( \lambda' \neq \lambda \)) scattering to just nearest-neighbor coupling (\( \lambda' = \lambda \pm 1 \)). In view of the translational symmetry, we are allowed to simulate carrier transport over the central —i.e., \( \lambda = 0 \) — stage only. Every time a carrier in state \( \nu \) undergoes an inter-stage scattering process (i.e., \( 0, \nu \rightarrow \pm 1, \nu' \)), the electron is properly re-injected into the central region (0, \( \nu \rightarrow 0, \nu' \)) and the corresponding electron charge \( \pm e \) will contribute to the current through the device (1). The proposed global-simulation scheme allows to study in a purely microscopic way —without resorting to phenomenological i/l parameters—the current-voltage characteristics of state-of-the-art unipolar nanostructures.

At this point a few comments are in order. Contrary to Eq. (1), the Boltzmann-like equation in (2) —describing an infinite system— does not contain explicit i/l terms, which are automatically accounted for within the proposed re-injection scheme. Moreover, the electric field does not enter our transport equation via the usual drift term (1); the corresponding bias is directly taken into account in evaluating the single-particle states \( \{kv\} \) (1).

It is well-known that the application of the present semiclassical transport theory to MQW structures at high fields becomes questionable. Indeed, according to (2), in the absence of scattering processes \( (P^{s}_{k\alpha,k'\alpha'} = 0) \) we have no current. This paradoxical result is due to the neglect of phase coherence between our localized states. A proper treatment of coherent transport —including resonant-tunneling effects— would require a density-matrix formalism (12). However, in the presence of a strong scattering dynamics current is mainly determined by incoherent hopping processes between localized states; moreover, such phase-breaking processes lead to a very fast decay of off-diagonal density-matrix elements. In this regime coherent-transport effects play a minor role and the semiclassical limit is recovered. It is then clear that the nature —coherent versus incoherent— of charge transport in QCLs is primarily dictated by the role played by incoherent scattering processes.

In order to discriminate between the two different regimes, we have applied the above global-simulation scheme to state-of-the-art QCL structures. In particular, as prototypical device, we have considered the diagonal-configuration QCL in (3), schematically depicted in Fig. 1, in which the proposed simulation scheme is also sketched. Here, the energy levels and probability densities of various electron states within the simulated stage \( (\lambda = 0) \) are also plotted: They correspond to the device active region (full line, \( \nu = 1, 2, 3 \)) as well as to the collector region (dashed line, \( \nu = A,B,C,D,E \) ). The nearest stages \( \lambda = \pm 1 \) are partially shown for clarity as well.

In order to properly model phase-breaking hopping processes, in addition to carrier-optical phonon scattering, all various intra- as well as intersubband carrier-carrier interaction mechanisms have been considered. To this end, the well-established MC method for the simulation of intercarrier scattering in quantum-well systems (3) has been extended to our MQW biased structure. Within such approach —based on the Born approximation—the effect of nonequilibrium multisubband screening of the bare two-body Coulomb potential is taken into account within the standard random-phase approximation (3). Other scattering mechanisms, not included in the simulation, are expected to play a very minor role (3): The interaction with acoustic phonons—which in the absence of carrier-carrier scattering would be essential to get ergodicity— does not affect energy relaxation significantly due to its quasi-elastic nature and small coupling constant; Carrier-impurity scattering is also expected to have very little impact on vertical-transport phenomena.

As a first step, we have investigated the relative weight of the carrier-carrier and carrier-phonon competing energy-relaxation channels. The time evolution of the carrier population in the various subbands as well as of the total current density in the presence of carrier-phonon scattering are depicted in Fig. 2. Parts (a) and (b) refer, respectively, to simulated experiments without and with two-body carrier-carrier scattering. In our “charge conservation” scheme, we start the simulation assuming the total number of carriers to be equally distributed among the different subbands; then the electron distribution functions evolve according to Eq. (2) and a steady-state condition is eventually reached, leading to the desired \( 3 \rightarrow 2 \) population inversion. As we can see, the inclusion of intercarrier scattering has signifi-
cant effects; It strongly increases inter-subband carrier redistribution, thus reducing the electron accumulation in the lowest energy level A and optimizing the coupling between active region and injector/collector (the populations of subbands 3 and B get equal). This effect comes out to be crucial in determining the electron flux through the MQW structure. Indeed, Fig. 2(c) shows the simulated current density across the core region of our QCL device, obtained with (full line) and without (dashed line) the inclusion of carrier-carrier scattering. These simulations have been performed at the threshold operating parameters estimated in [13]. The current density in the presence of both electron-phonon and electron-electron scattering mechanisms is about 4 kA/cm²: This value is in good agreement with the experimental results in [13].

The results plotted in Fig. 2 clearly demonstrate that the electron-phonon interaction alone is not able to efficiently couple the injector subbands to the active region ones: While carrier-phonon relaxation well describes the electronic quantum cascade within the bare active region [6], carrier-carrier scattering plays an essential role in determining charge transport through the full core region. This can be ascribed to two typical features of carrier-carrier interaction — compared to the case of carrier-phonon —: (i) this is a long-range two-body interaction mechanism, which also couples non-overlapping single-particle states [see Fig. 1]; (ii) the corresponding scattering process at relatively low carrier density is quasielastic, thus coupling nearly resonant energy levels, like states 3 and B.

This result is extremely important: it tells us that the presence of a coherent-tunneling dynamics — often invoked to account for nearly resonant inter-level processes between active region and injector/collector — is not necessary: the charge transport in QCL structures can be explained quantitatively in terms of incoherent intercarrier-assisted tunneling processes, and thus within a purely semiclassical transport theory. This allows us to answer the long-standing controversial question on the nature — coherent versus incoherent — of charge transport in QCLs previously mentioned: for the typical structures considered, energy-relaxation and dephasing processes are so strong to destroy any phase-coherence effect; as a result, the usual semiclassical description is in excellent agreement with experimental results.

Since in the proposed MC simulation scheme, the current density across the whole structure is an output of the simulation, the current-versus-voltage behavior of our semiconductor quantum device can be obtained within our fully microscopic description, without resorting to any phenomenological external parameter. Figure 3 presents the simulated voltage-current characteristics for the MQW structure of Fig. 1. As we can see, the current density increases with the applied field, due to the increased coupling between subband 3 and various lower-energy injector states, in which carriers tend to relax and accumulate. The agreement between our theoretical results and the measured dependence shown in Ref. [13] is extremely good. Discrepancies between simulated and measured values are well within the measurement uncertainties and the complexity of the real device structure.

In summary, we have presented the first global simulation of semiconductor-based QCLs. The proposed theoretical scheme allows for a fully microscopic evaluation of the current-voltage characteristics of prototypical QCL structures. The excellent agreement between our semiclassical simulations and recent experimental results confirms the incoherent nature of charge transport in such unipolar quantum devices.

This work has been supported in part by the European Commission through the Brite Euram UNISEL project (Contract No. BE97-4072).

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[15] The apparent discrepancy between the experimental ($\approx 7 \text{ kA/cm}^2$) and the theoretical values ($\approx 4 \text{ kA/cm}^2$) is due, we believe, to the different estimate of the potential drop per period required to line up the ground state A of the injector with level 3 of the active region. Our single particle calculation, which takes into account Poisson renormalization of the band structure, allows to predict a good alignment at a field of 68 kV/cm. This corresponds to a potential drop of 9.2 V along the device thirty-period core region, in much better agreement with the measured value of 11.4 V than the 6.9 V at a field of 48 kV/cm quoted in Ref. [13]. As shown in Fig. 2(c) the simulated current density at 68 kV/cm is about 7 kA/cm$^2$, thus enforcing the present interpretation.

FIG. 1. Schematic representation of the conduction-band profile along the growth direction for the diagonal-configuration QCL structure of Ref. [13]. The MQW is biased by an electric field of 48 kV/cm. The levels $\nu = 1, 2, 3$ and $\nu = A, B, C, D, E$ in the active and collector regions (full and dashed lines, respectively) of the simulated stage ($\lambda = 0$) are also plotted together with the corresponding probability densities. The replica of level 3 in the following stage $\lambda = +1$ is shown for clarity.

FIG. 2. Time evolution of simulated carrier densities in the various subbands of the MQW structure of Fig. 1 (full lines: $\nu = A, B, C, D, E$; dotted lines: $\nu = 1, 2, 3$), without (a) and with (b) intercarrier scattering. (c): current density across the whole structure in presence (full line) and absence (dotted line) of carrier-carrier interaction. An electric field of 48 kV/cm has been applied.

FIG. 3. Applied-field vs current-density characteristic at 77K of the MQW structure of Fig. 1. The dashed line is a guide to the eye.
R. C. Iotti and F. Rossi, Figure 1
R. C. Iotti and F. Rossi, Figure 2(a)
R. C. Iotti and F. Rossi, Figure 2(b)
R. C. Iotti and F. Rossi, Figure 2(c)
R. C. Iotti and F. Rossi, Figure 3