

4.13 Report of project part 13: Mesoscopic theory of quantum devices

Theory of optical properties and carrier transport of mid-infrared quantum-dot devices

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4.13.1 Summary

The aim of the of this theory project was to develop electronic structure and quantum transport methods that are optimally suited for the mesoscopic length scale of nanostructures and apply them to the mid-infrared device structures relevant for this collaborative project. Consequently, we primarily pursued three research directions, in full accord with our original application:

- develop a new, computationally efficient and modular version of the nextnano simulation suite to be able to predict the electronic and optical properties of realistic, embedded 3D quantum dot structures.
- develop a fully self-consistent non-equilibrium Green's function method to be able to faithfully predict and help designing and optimizing THz quantum cascade structures.
- develop a spin-dependent non-equilibrium Green's function method to study spin-dependent transport in 2DEGs.

We have carried out all three projects successfully and have reached a level that enables us to calculate and analyze sophisticated and highly complex nanostructures such as coupled quantum dot layers embedded in multi quantum wells, novel QCL designs, or optical absorption spectra of piezoelectric quantum dots of various shapes.

In total, we have published 32 papers in conjunction with this project that financially supported one PhD student for 3 years and a second PhD student for the last year. Formally, there was 1 postdoc and 1 PhD student granted for the project but the state-tariffs in Bavaria, Germany, covered only the costs of 1.3 PhD student positions.

While we have started several intense collaborations with P2, P3, P7, P11, P12, only a few papers [Brehm 2008, Grützmacher 2007] have already appeared in print up to now but several more have either been submitted or are in the pipeline of being submitted.

4.13.2 Scientific Background – State-of-the-Art

4.13.2.1 Theory of electronic structure in semiconducting nanostructures

Electronic structure calculations in semiconductors are either atomistic or based on a continuum approach (see reviews Majewski 2004, Kümmel 2008). Among the atomistic methods, there are ab-initio methods based on density functional theory and semi-empirical methods such as tight-binding approaches. The latter approach is semi-quantitative at best but yields faithful chemical trends and provides a good physical insight. Ab-initio methods are the most reliable, predictive schemes available yet suffer from two drawbacks. First, the computational effort limits these methods to a few hundred or thousand atoms. Secondly,

these methods are accurate on the energy scale of Rydberg but not on milli-Rydberg which is, in many cases, the relevant energy scale in semiconductors. If one seeks a global picture of the electronic structure and optical properties of semiconductor nanostructures on a mesoscopic scale, the method of choice is the effective mass theory that generalizes the k.p method (Ivchenko 1997, Foreman 1997). There are several variants of this method but all of them share the same limitations. A continuum approach is bound to fail if a system changes significantly on an atomic scale. In practice, most semiconductor nanostructures are alloys and show a fairly smooth transition in their electronic structure so that this approach works reasonably well down to structures of a few nm.

This project has focused on the understanding of nanostructures on a mesoscopic scale and therefore used the effective mass approach. However, we have generalized these methods significantly to ensure reliable, robust and physically meaningful results. These will be further discussed below.

4.13.2.2 Theory of carrier dynamics in quantum cascade lasers

The carrier dynamics in QCLs is commonly estimated by separating the calculation of the energy states and their occupation. The former are calculated by the single-particle Schrödinger equation, and the semiclassical Boltzmann equation or a simple rate equation is used to determine the occupation of states. This procedure obviously misses the finite width of the resonant electronic states, their significant change with occupation and all coherent tunneling effects that couple more than adjacent quantum wells. Thus, this procedure is only useful for qualitative considerations.

The most elaborate formalism that captures both quantum mechanics and nonequilibrium occupation of states in a consistent fashion is the Keldysh or nonequilibrium Green's function formalism (NEGF). The first elaborate adaptation of the Keldysh formalism to semiconductors is due to Lake and coworkers (Lake 1997). Due to computational limitations at the time, the authors were forced to use several simplifications in their concrete applications but this seminal work is still an invaluable source for most scattering self-energies in semiconductors. This formalism was applied to QCLs very extensively by Wacker and coworkers (Wacker 2002). Basically, these papers discuss all electronic aspects of QCLs, including the evaluation of optical spectra.

A problem that has plagued all published NEGF calculations in this context is that this formalism does not automatically conserve important physical observables such as charge, current or the Pauli exclusion principle. The use of Wannier-type basis functions by Wacker, for example, typically leads to a 10% variance of the current throughout the device (Wacker

2002). This may be acceptable for some devices, but it is clear that a reliable and robust method should always guarantee physically correct results.

As will be discussed in the next section, we have implemented a fully self-consistent NEGF approach that guarantees charge, current conservation and the Pauli principle automatically and includes the complete spatial, momentum and energy dependence of the scattering self-energies. This accuracy has turned out to be vital for reliable predictions and interpretations of the QCL structures that have been fabricated in this collaborate project.

4.13.3 Results and Discussion

4.13.3.1 Nextnano++

Nextnano is a simulation software suite for the charge-self-consistent solution of the multi-band k.p-Schrödinger-Poisson equation in arbitrarily shaped and strained three-dimensional (3-D) nanostructures and for the calculation of ballistic or diffusive carrier transport. We have developed this package over the last 10 years. When this collaborative project started, nextnano had grown to a Fortran 90 code base of more than 200000 lines that had become unmanageable. Devices of effectively 1, 2, or 3 dimensions were treated independently so that a bug needed to be fixed in several code locations. Importantly, there were several open issues in the physical modeling, in the mathematical implementation, and in the code structure.

We have rewritten nextnano from scratch in C++, adhered to strictly object oriented, modular coding, generalized or newly developed almost all physical models and finally were able to accelerate execution times by a factor of 10 to 50, significantly widening the scope of problems that can be tackled successfully [Birner 2007, Trellakis 2007, Trellakis 2006a, Birner 2006, Trellakis 2006b]. We have named the new version nextnano++ and it is freely available on our web site.

The electronic structure is calculated in terms of an 8-band effective mass theory in nextnano++. We have been able to eliminate the spurious solutions and related artifacts that have plagued effective mass approaches previously by properly symmetrizing the Hamiltonian and ensuring that it is not only Hermitian but manifestly self-adjoint. In addition, we have developed a method that allows us to treat gap-less materials such as type-II heterostructures or doping superlattices with large variations in the band gap. We have significantly generalized the calculation of optical absorption within the effective mass scheme and can now reliably calculate both intraband (inter-subband) as well as interband optical transition probabilities. Finally, we have developed a novel method to calculate magnetic fields effects in a manifestly gauge invariant manner [Trellakis 2006a].

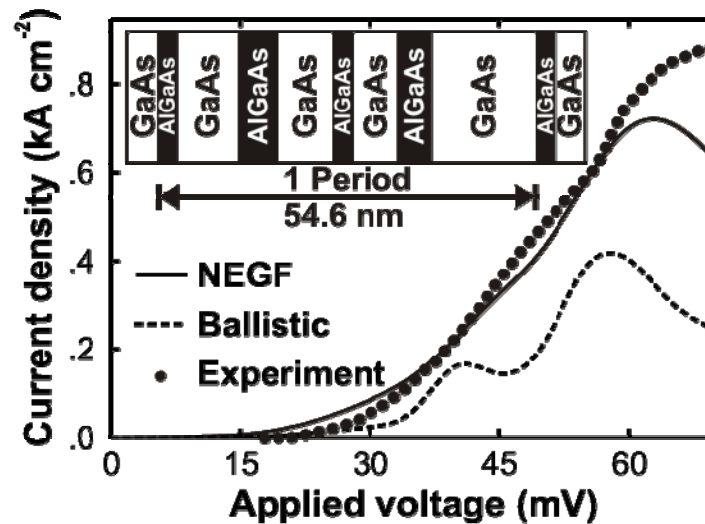
We have carefully assessed the strengths and weaknesses of nextnano++. Effective mass theory is a continuum approach and therefore gradually fails for strongly confined systems when the spatial extent of the wave functions approaches less than 5 nm. On the other hand, effective mass theory enables one to realistically include long-range Coulomb charging and elastic strain effects which is still out of reach for ab-initio approaches. We have systematically studied nitride quantum dots and other nanostructures in order to assess the accuracy of the calculated piezoelectric and pyroelectric polarization charges [Lee 2006a, Lee 2006b, Galluppi 2005, Povolotskyi 2004a, Povolotskyi 2004b, Povolotskyi 2004c].

Among the most successful applications of nextnano++ were the predictions of excitonic spectra in electronically coupled vertically stacked self-assembled InGaAs quantum dots [Nakaoka 2006, Krenner 2005, Finley 2004, Finley 2004]. We have been able to quantitatively explain the cross-over from indirect excitons, where electron and hole sit on different quantum dots, to excitons where the hole sits on one dot and the electron forms a bonding and an antibonding state, respectively.

In collaboration with P7 and P12 and with experimental groups at TU Munich, we have successfully studied the mesoscopic electronic structure SiGe dots [Brehm 2008, Grützmaier 2007, Bougeard 2004] that helped optimizing the optical properties of these structures.

4.13.3.2 THz Quantum Cascade Lasers

For theorists, the electronic carrier dynamics in quantum cascade lasers (QCLs) poses a significant challenge. The electronic transport depends on scattering by phonons and therefore requires the incorporation of scattering. On the same time, the electronic transport utilizes resonant tunneling and quantum mechanical interference. Either effect by itself is relatively easy to cope with: incoherent scattering can be calculated in terms of the Boltzmann equation and resonant tunneling is a feature of ballistic transport that can be evaluated by solving the Schrödinger equation with open boundary conditions but ignoring any kind of incoherence. By contrast, a realistic theory of the carrier dynamics in QCLs requires one to treat quantum mechanics and incoherent scattering mechanisms on an equal footing. Basically, quantum mechanics provides the electronic states and the scattering mechanisms determine the occupation of these states. The method of choice that includes both effects consistently with one another is the Keldysh Green's function approach, usually referred to as nonequilibrium Green's function theory (NEGF).



We have developed and implemented what we believe to be the most accurate and fully self-consistent NEGF theory for QCLs to date [Birner 2008, Kubis 2008a, Kubis 2008b, Kubis 2007, Kubis 2005]. By utilizing novel and computational techniques and implementing them efficiently, we have been able to eliminate most simplifying assumptions pertaining to the momentum and energy dependence of scattering mechanisms that have been invoked previously. Our method takes into account the spatial nonlocality of all relevant incoherent scattering mechanisms, i.e. acoustic, polar-optical phonon scattering, impurity, Hartree electron-electron, and interface roughness scattering. This guarantees realistic scattering rates as well as accurate state energies, lifetimes, and state occupations. The optical gain is calculated in terms of linear optical response, taking into account the calculated non-equilibrium stationary state occupations. The present theory does not contain any adjustable parameters. Instead, all coupling constants that enter the model are well established material parameters.

We have systematically studied GaAs/AlGaAs based THz-QCL structures that have been fabricated recently in project P11 as a function of doping, and lattice temperature, interface roughness. Our results for the I-V-characteristics, for the threshold current, and for the emission frequency show excellent agreement with experiment. This is exemplified in the figure on this page that shows the I-V curve of a GaAs/Al_{0.15}Ga_{0.85}As QCL at 40K and compares the present NEGF calculations (full line) with a strictly ballistic calculation that ignores scattering altogether (dashed) and with experiment (dots, see project P11), respectively. The calculations show that

- Stationary charge transport in THz-QCLs is dominated by quantum mechanical multi-barrier tunneling at low bias rather than by sequential tunneling.

- Interface roughness tends to increase the current density significantly due to the relaxed lateral momentum conservation. We also find, however, that rough interfaces suppress the gain drastically.
- Increasing the lattice temperature from approximately 5 K to 80 K increases the current density by one order of magnitude. For higher temperatures, a mismatch of the carrier distribution with tunneling resonances actually leads to a reduction of the current.
- The threshold current increases linearly by increasing the collector sheet doping density up to doping concentrations of $2 \times 10^{10} \text{ cm}^{-2}$. For higher doping levels, we predict a superlinear increase of the threshold current that agrees excellently with recent experiments.

While our model contains no fitting parameters, it does contain quantities that are not accurately known, such as the effective lattice temperature inside the active region and the interface roughness. It turns out that we can only get agreement with all experiments in very narrow windows for these parameters which gives an important feedback to experiment.

4.13.3.3 Spin Polarization

In accordance with our proposal, we have developed and implemented a spin-dependent NEGF theory for transport through nanostructures. In particular, we have studied the intrinsic spin Hall effect as a function of geometry of laterally confined 2D electron systems. Interestingly, we have found that the spin-orbit interaction in a p-doped GaAs 2DEG leads to a significant spin polarization for a 3-terminal, T-shaped nanostructure [Kubis 2008c].

Since, however, the experimental groups in this collaborative project have not focused on spin-dependent effects, this part of our project will not be further pursued in this framework.

4.13.4 Collaboration within and beyond the SFB

In this first period of the collaborative project, we have focused on the development of theoretical tools such as `nextnano++` and the NEGF method. While these codes will hardly ever reach a “final” state, they already enabled us to carry out intense collaborations with the projects P2 and P12 on the mesoscopic electronic structure of SiGe dots, aided by the x-ray experiments of P7. Similarly, we carried out several systematic studies of the optical properties and transport characteristics of GaAs/InGaAs multi quantum wells and quantum cascade structures and of self-assembled quantum dots in collaboration with P3, P11. Several papers have already been published with these groups but the highly fruitful collaboration with P3 and P11 on QCLs and quantum dots will lead to many more joint publications in the near future.

The development of a large software package such as nextnano requires expertise in many different areas in physics, numerical mathematics, and computer science. Its usefulness depends critically on the feedback and application by many theoretical and experimental groups. Indeed, we have collaborated with many colleagues worldwide, based on results obtained with the new version of nextnano++. In particular, we have published joint papers with colleagues from the Universities of Modena (Italy), Purdue (US), Tokyo (Japan), ASU in Phoenix (US), Sheffield (UK), Rome (Tor Vergata, Italy).

In developing our NEGF method for the study of QCLs, we were fortunate to get the continuous help of Andreas Wacker from Lund (Sweden). His expertise in lasers systems in general, and his deep knowledge of the NEGF technique played an essential role in our progress. We intend to deepen and extend our collaboration with him in the future.

4.13.5 References

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