Nano-electronics industry has during the past decade decreased feature sizes to roughly 10nm. Such feature sizes are at the quantum limit, requiring a description at the quantum mechanical level. Parallel to the experimental work reside the theoretical tools used to investigate and understand such systems. These theoretical tools still have a high computational requirement. Thus more efficient algorithms are needed to perform studies on even larger systems. Although the gap between the theoretical tools and the experimental setups are reduced, there is still a gap, and the used theoretical methods require revised algorithms.

Furthermore, the advent of 2D materials may prove prominent in future nanoelectronics for electronic and heat transport devices. Such materials include the Nobel Prize winning material, graphene which has unique properties.

The main focus of the work presented in this thesis has been to introduce extensions to the non-equilibrium Green function code TranSIESTA which will help reduce the gap between experimental and theoretical studies. One main achievement in this work is a truly $N_e \geq 1$ electrode implementation. Another contribution is a general $N_e \geq 1$ tight-binding transport code which enables not only electronic transport but also phonon transport. This tight-binding code includes features such as, bond-currents, transmission projections and bias-interpolations. For both codes the inversion algorithm for calculating the Green function is revised. We implement a high performing block-tridiagonal algorithm for calculating the Green function and spectral density function — even for $N_e > 2$. This is accomplished by bandwidth reduction schemes that increase the quasi-1D interpretation.

We also present a new gating method capable of calculating gating effects. A graphite example is used to highlight the importance of the quantum capacitance that is evident in low density of states systems. Additionally the gating method was used in nonequilibrium to study the gate-bias dependence on graphene nano-constrictions. This indicated a pinning effect arising due to differences in coupling strength between the device and the two electrodes.

Two studies are presented using the non-equilibrium method with $N_e = 3$. First, graphene T-junctions are studied to uncover potential interconnects in future graphene based devices. This T-junction is studied under two non-equilibrium situations. Our second study is a graphene Scanning Tunnelling Microscopy setup where the inelastic current is calculated. We show, that the experimental inelastic signal may be fully recovered using density functional theory and non-equilibrium Green function techniques.