Scientific Report "Short Visit Grant"

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Purpose of the visit: Prof. Robert van Leeuwen and myself published several works on time-dependent phenomena at the nanoscale using the nonequilibrium Green's function approach. This approach is based on the solution of the so called Kadanoff-Baym equations which are nonlinear integro-differential equations for the Green's function. Due to their consistent computational cost we are exploring suitable approximations to extend the maximum propagation time and be able to perform ab-initio simulations of charge transfer processes occurring either between two molecules or between a molecule and a solid. We are currently involved in setting up a code which implement an approximate Kadanoff-Baym dynamics. This requires a continuous benchmark with the full dynamics in order to establish range of applicability and limitations. The purpose of the visit was to assess the quality of the approximate dynamics in simple donor-acceptor complexes by testing different implementation schemes, self-energies, correlation regimes, etc.

Description of the work carried out during the visit: We used a tight-binding Hamiltonian to describe a donor and a chain of acceptor sites. This donor-acceptor complex was then sandwich between two metallic leads and driven out of equilibrium by a radiation field. We calculated the local densities and currents for different coupling to the leads, interaction strengths, and donor-acceptor transition rates using different self-energies and implementation schemes.

Description of main results obtained: We carried out the first simulations and benchmarks. The approximate dynamics was in a remarkable good agreement with the full Kadanoff-Baym dynamics at the Hartree-Fock level for all cases considered. We discussed self-energy approximations in order to include correlation effects. This is a delicate issue in the approximate scheme since correlation effects enter in the quasi-particle dynamics and in the collision integrals in a separate manner. Among the various implementation schemes we could find one which reproduces several features of the full Kadanoff-Baym dynamics. This holds promise for incorporating correlation effects in a time-dependent Green's function framework which scales only quadratically with the propagation time.

Future collaboration with the host institution: We will continue in developing the approximate Kadanoff-Baym scheme. There are several things which are currently missing like, e.g., the inclusion of screening and the electron-phonon interaction.

Projected publications: We are currently writing a draft in which a detailed derivation of the approximate scheme is discussed and the first comparisons with a full Kadanoff-Baym dynamics are presented.