A State Representation Approach for Atomistic Time-Dependent Transport Calculations in Molecular Junctions

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A new method for simulating electron dynamics in open quantum systems out of equilibrium, which is motivated by the intuitive and practical nature of the damped Liouville von-Neumann equation approach of Sánchez *et al*. [J. Chem Phys, 124, 214708 (2006)], will be presented. The new approach is based on a transformation of the Hamiltonian matrix from an atomistic to a state representation of the molecular junction. This enables us to uniquely define the bias voltage across the system while maintaining a proper thermal distribution within the lead models. Furthermore, it allows us to investigate time-dependent effects in non-linear and multi-lead configurations. We investigate the degree of conservation of exact conditions such as the *N*-representability and positivity of the density matrix. We believe that the new approach offers a practical and physically sound route for performing atomistic time-dependent transport calculations in realistic models of molecular electronics junctions.