Time-Linear Quantum Transport Simulations: Electroluminescence rectification and high harmonic generation in molecular junctions

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Molecular systems are prospective elements of future electronic devices such as nano-junctions. State-of-the-art calculations are able to accurately predict ground and excited properties of technologically relevant molecules. However, *ab initio* description of photo-assisted tunneling, optical rectification, and electrically driven photon emission requires a new set of tools.

Therefore, I present a time-linear scaling method to simulate open and correlated quantum systems out of equilibrium [1]. The many-body diagrammatic theory provides a systematic approach to handle interactions between electrons, bosonic excitations, and embedding. To access the dynamical properties of the system, the Kadanoff-Baym equations for the two-time electron and boson Green's functions must be propagated. The time nonlocality of the scattering term poses a significant challenge for full two-time propagation, resulting in at least cubic scaling with the physical propagation time. The generalized Kadanoff-Baym ansatz (GKBA) alleviates the scaling problem by limiting the propagation to the time diagonal and working with density matrices rather than Green's functions. This approach leads to a coupled system of first-order ordinary differential equations (ODEs) with linear time scaling, as demonstrated in electronic [2], electron-bosonic [3], and open systems within the wide band limit approximation (WBLA) [1].

As a case study for the formalism, I report the quantum pump effect in a Benzenedithiol molecule connected to copper electrodes and coupled with cavity photons [4]. The nonequilibrium transport simulations by the recently developed Cheers code [5] reveal electric and photonic current responses to an ac bias voltage, pronounced electroluminescence and high harmonic generation (HHG) in this setup. The mechanism of HHG is more analogous to that from solids than from isolated molecules. Comparing the power carried by the photon flux with the total power, we found quantum efficiency around ten percent, similar to quantum-dot devices.

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