Nonequilibrium steady-states in photodoped Mott insulators

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Photodoped states in Mott insulators are peculiar states which simultaneously host strongly correlated electron and hole-like carriers, and can show instabilities into various non-thermal orders. Here we stabilize a stationary photodoped state in a Mott insulator using Dynamical-Mean-Field-Theory (DMFT) in the nonequilibrium steady-state formalism. This formalism provides a description of the longtime dynamics of microscopic models with well separated timescales. The photodoping can be established by coupling the Hubbard model with external baths that pump holon and doublon excitations in the Hubbard bands. In particular, we develop an algorithm to stabilize DMFT solutions with a prescribed nonthermal distribution function in the local Green's functions. This formulation may allow for a nonperturbative solution of the DMFT impurity model, using methods like Quantum Monte Carlo, and it opens the possibility to study the dynamics of photodoped states using a Quantum Boltzmann equation.

Thermalization of a beam of positive charge carriers in a quasi-1D electron gas

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Quantum plasmas in nonequilibrium are of growing interest in recent decades. A reliable description of quantum phenomena as well as dynamical screening and strong coupling is required to resolve these exotic systems. These high demands are fulfilled by quantum kinetic approaches [1] such as NEGF.

Classical NEGF calculations suffer from severe drawbacks due to their N_t^3 CPU scaling with the propagation time N_t, which stays true even with the Generalized Kadanoff-Baym Ansatz (GKBA) [2] for most selfenergy approximations. The recently developed G1--G2 scheme [3,4], a time-local reformulation of the GKBA with a CPU scaling of N_t, offers the possibility to reach very long propagation times and for a variety of selfenergies, which are needed e.g. to reach good spectral resolution.

We now use this method to model a uniform quantum plasma. Due to the large dimension of the two-particle NEGF introduced in the G1—G2 scheme, only a quasi-1D quantum wire geometry is currently feasible. After earlier applications using a statically screened Second-Order-Approximation (SOA) [5], we now extend our studies to GW selfenergies and thereby including dynamical screening as well as nonequilibrium plasmon dynamics.

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Towards calculating intersystem crossings in extended systems implying periodic boundary conditions

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One major problem in fighting climate change is the production of a sufficient amount of energy while remaining at competitive cost without emitting CO2 or other greenhouse gases. Therefore, more efficient ways to produce electricity using green technology are needed. One approach to increase the efficiency of solar panels is by introducing singlet fission materials into a single-junction to generate multiple low energy excitons from a single excitation, thus increasing the useable part of the electromagnetic spectrum[1]. To aid this development with quantum chemical calculations, efficient methods for calculating spin-orbit coupling and intersystem crossing within the condensed phase are needed. We present the first implementation of perturbational spin-orbit couplings for all-electron computations within the restricted closed-shell time-dependent density functional theory (TDDFT) module of the CP2K program package[2-4]. The perturbative ansatz enables the construction of both singlet and triplet contributions relying on an ”auxiliary” set of many-electron wavefunctions[5]. Our code within the TDDFT module is based on and checked against the related module for X-ray absorption spectroscopy[6]. The reported method utilizes the already established TDDFT implementation of CP2K together with the quasi-relatistic ”Zeroth Order Regular Approximation” (ZORA) Hamiltonian[7,8]. We will show absorption spectra for various small molecules as well as selected cer-based metal-organic frameworks

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Energy flow during relaxation in an electron-phonon system with multiple modes

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One of the important goals of nonequilibrium condensed matter physics is to understand the relaxation dynamics. Theoretical studies using the multi-temperature model (MTM) or the Boltzmann equation (BE) have pointed out complex relaxation processes involving couplings between electrons and multiple phonon modes. One example is the backward energy flow, where the direction of energy flow between electrons and certain phonon modes is reversed during the relaxation process. This phenomenon can be intuitively understood as an inversion of relation between the electron and phonon effective temperatures using MTM. However, many approximations are used to derive BE and MTM from the nonequilibrium Green's function method (NEGF). Therefore, it is important to go back to NEGF and discuss the validity of the picture provided by MTM and BE for complex energy flows.

In this study, we introduce an extended Holstein model where electrons couple to two phonon modes, and analyze this model using the nonequilibrium dynamical mean-field theory. We derive approximated energy flow equations related to BE and MTM, and compare them to the full energy flows obtained from NEGF. In the weak electron-phonon (el-ph) coupling regime, we find that the inversion of relation between two nonequilibrium distribution functions of Green's functions and self-energies leads to the backward energy flow. Since the temperatures derived from the distributions of phonon Green's function and self-energy are closely related to the effective temperatures of phonon and electron in MTM, we consider that the interpretation of the backward flow using MTM is applicable. On the other hand, in the strong el-ph coupling regime, we find that the origin of energy flow cannot be explained solely by nonequilibrium distribution functions. This indicates the importance of conducting an analysis of the full energy flow derived from NEGF in the strong el-ph coupling regime.

Universal approach to non-adiabatic dynamics of finite and extended atomistic systems

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We recently proposed a framework for non-adiabatic dynamics under general non-equilibrium conditions [1] based on an infinite hierarchy of equations of motion (EoM) for various combinations of positions and momenta of atoms. A real-time evolution is naturally coupled to the thermalization of the system. Stochastic EoM for atoms are derived based on integrating out electronic degrees of freedom via path integrals and deriving a stochastic Liouville equation for the ionic reduced density matrix. Complex colour noises are used based on correlation functions expressed via non-equilibrium Green's functions (NEGF) of the electronic subsystem. The physically significant trajectories are obtained by sampling over all stochastic trajectories.
In this paper we propose a general technique of building an arbitrary level of hierarchy of EoM employing an interplay of Weyl quantization and Wigner transformation methods [2]. This opens up a possibility of running the hierarchy of EoM until convergence, at least in principle.
The second point considered here is related to a general method of generating complex multi-variable colour noises based on the correlation functions due to electrons. Our method is based on a generalization of an existing algorithm [3] based on circular embedding and toeplitz matrices.
The developed method is applied to a simple single-site single-mode model quantum atom in a quantum junction (SSSM).
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A NEGF-GKBA approach to second harmonic generation in the Dicke's model.

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Due to their conceptual and computational convenience, few-level systems are often studied to investigate fundamental aspects of nonlinear optical behavior, for example second harmonic generation (SHG). Here we focus on the low-photon regime of SHG, where quantum effects due to the radiation field can play an important role [1], and use as system of interest the Dicke's model [2]. In its standard form, the model consists of several two-level systems (TLS) interacting with one photon mode. To study SHG, we augment the model with a second (the fluorescent) mode, and at the same time adopt a real-time description of SHG [3]. The Dicke's model is usually studied in the homogeneous limit, i.e. with identical TLS, but it is also of interest to consider the case where disorder and/or electron-electron interactions are present [4]. In this work, we address this situation in large Dicke's systems by considering the nonequilibrium Green's function (NEGF) method within the Generalized Kadanoff-Baym Ansatz (GKBA)[5]. Our preliminary results show that both disorder and interaction tend to reduce the intensity of the fluorescent spectra in time while, not unexpectedly, increasing the number of TLS (i.e. the system's size) generally enhances the optical response.

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Quantum chemical calculation for photo-absorption spectra in solution using reference interaction site model

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Simulating the absorption spectrum line of solutions under the visible light region is a challenging topic in computational and physical chemistry. This is mainly due to the difficulty in dealing with the mol orders degrees of freedom of solvent molecules and quantal fluctuations of a target molecule. We present a hybrid approach that combines Green's function and linear response theory for the target molecule and reference interaction site model (RISM), one of the molecular liquid state theories, for the solvent. Our methodology is constructed based on statistical mechanics and only uses microscopic-interaction. The unperturbed effective Hamiltonian of the solute molecule is corrected by incorporating the solvation effect using RISM, and the perturbation term is given by the sum of dielectric coupling between light and charged particle and solvation energy deviated from the equilibrium configurations of solvents. We obtained the damping function causing quantal decoherence of the vibration of solute by applying the randomness of the solvent configuration and fluctuation and dissipation theorem. Notably, our method does not include any empirical or artificial parameters in simulating the spectral line. We numerically demonstrated the 2-Thiocytosine keto form, one of the analogue of nucleic-acid, and the spectral band fits the experimental data very well. Our approach can be useful in studying the behavior of molecules in solution, and the obtained results can be compared with experimental data.

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Influence of phononic dissipation on impact ionization pro- cesses in a photodriven Mott insulator

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It has been suggested that in strongly correlated materials, highly photoexcited charge carriers could use their extra energy to excite additional carriers across the Mott gap via impact ionization [1,2]. However, the influence of electron-phonon scattering on photocurrent and impact ionization in Mott photovoltaic setups is still an open question.
We address this issue in a nonequilibrium steady state study on the occurrence of impact ionization in a simplified model of a Mott photovoltaic device in presence of acoustic phonons [3], consisting of a Mott-insulating layer coupled to two wide-band fermion leads. For a small hybridization to the leads, we obtain a peak in the photocurrent as a function of the driving frequency which can be associated with impact ionization processes, while for larger hybridizations we find a suppression of impact ionization with respect to direct photovoltaic excitations. The effect of acoustic phonons produces a slight enhancement of the photocurrent for small driving frequencies and a suppression at frequencies around the main peak at all considered hybridization strengths.

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Correlated Mott insulators in a strong electric field: The effects of phonon renormalization

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We characterize the response of a Mott insulating system to a static electric field in terms of its conducting and spectral properties. Dissipation is included by a coupling to fermionic baths and to either optical or acoustic phonons. This paper extends and completes the analysis made in a previous work by the authors [Mazzocchi *et al.*, Phys. Rev. B **106**, 125123 (2022)]. In the present work, phonons are included *diagrammatically* within the Migdal approximation by also including self-consistency from the electronic feedback. The nonequilibrium steady state is addressed by means of the dynamical mean-field theory based on the nonequilibrium Green's function approach, while the so-called auxiliary master equation approach is employed as impurity solver. With optical phonons the self-consistency suppresses the steady-state current for field strengths comparable to the band gap with respect to the non-self-consistent case. This is due to the interaction of phonons with the hot electrons of the lattice which increases their temperature, thus providing a less effective relaxation channel for the current-induced Joule heat. In addition, in the case of optical phonons, the results are essentially independent of the temperature of the fermionic baths, as the latter is sensibly smaller than their characteristic frequency. On the other hand, with acoustic phonons the steady-state current is slightly suppressed by the self-consistent treatment only at field strengths close to half of the gap and especially at very small phonon frequency. Also, in this case, the results seem to slightly depend on the temperature of the fermionic baths.

Multiphoton Photoemission and Floquet Dressing on Metal Surfaces

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Simple metal surfaces such as Cu(111) or Ag(111) are considered as benchmarks for photoemission spectroscopy with nonlinear optical responses. Using multiphoton photoemission, dressed Shockley surface states (SS) and image potential (IP) states can be experimentally probed, which makes such metal surfaces good platform for investigating Floquet physics.

Starting from mean field model potential, we utilize time-dependent surface flux method and Floquet non-equilibrium steady state calculation (NESS) to simulate interferometric-time-resolved multiphoton photoemission (ITR-mPP) spectrum. Our calculation successfully reproduces previous experimental data, and explains the splitting features with Floquet side bands. Furthermore, Fourier analysis of the ITR-mpp spectrum shows phase shift in different Floquet bands, which we found can be related to time difference between avoided crossing points in instantaneous Floquet state (IFS) quasienergy spectrum. At these points, population transfer from occupied surface states to Volkov states via Landau-Zener transition. Our finding suggests that nonlinear photoemission with strong field can be understood with the help of Flouquet picture.

Dynamical exchange-correlation potential formalism in spin$-\frac{1}{2}$ systems with Heisenberg and Dzyaloshinskii–Moriya interactions

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We present an exchange-correlation potential formalism, tailored for spin Hamiltonians, which extends to the spin-only case a formulation previously introduced and applied to the one-dimensional Hubbard model. Within the spin exchange-correlation potential formulation, a new sum rule for spin-systems is derived. We apply the formalism to spin$-\frac{1}{2}$ systems with competing Heisenberg and Dzyaloshinskii–Moriya (DM) interactions, and in the presence of an external magnetic field. For the case of a spin-chain, the exchange-correlation potential in the thermodynamic limit is extrapolated from exact diagonalization/density matrix renormalization group results of small clusters. For a purely antiferromagnetic interaction, comparisons with numerical benchmarks show that the extrapolation yields a one-particle spectral function with favourable accuracy at a relatively low computational cost. These results are contrasted to the case where also DM interactions are present, and preliminary results for two-dimensional finite systems are also reported. Finally, we are currently applying the formalism to non-equilibrium system and results will be shown depending on the status of progress at the time of the workshop.