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THE TIME-DEPENDET QUANTUM TRANSPORT IN NANOSYSTEMS

We propose a time-dependent many-body approach to study the short-time dynamics of correlated electrons in quantum transport through nanoscale systems contacted to metallic leads. This approach is based on the time propagation of the Kadanoff-Baym equations for the nonequilibrium many-body Green's function of open and interacting systems out of equilibrium.

1. Introduction

The description of electron transport through nanoscale systems contacted to metallic leads is currently under intensive investigation especially due to the possibility of miniaturizing integrated devices in electrical circuits.Several theoretical methods have been proposed to address the steadystate properties of these systems.

Ab initio formulations based on time-dependent (TD) density functional theory (see [1]) (DFT) and current density functional theory [2-4] provide an exact framework to account for correlation effects both in the leads and the device but lack a systematic route to improve the level of the approximations. *Ad-hoc* approximations have been successfully implemented to describe qualitative features of the $I(V)$ characteristic of molecular junctions in the Coulomb blockade regime.

The possibility of including relevant physical processes through an insightful selection of Feynman diagrams is the main advantage of many-body perturbation theory (MBPT) over one-particle schemes. Even though computationally more expensive MBPT offers an invaluable tool to quantify the effects of electron correlations by analyzing, e.g., the quasiparticle spectra, lifetimes, screened interactions, etc. One of the most remarkable advances in the MBPT formulation of electron transport was given by Meir and Wingreen who provided an equation for the steady-state current through a correlated device region thus generalizing the Landauer formula.(see [3]). The Meir-Wingreen formula is cast terms of the interacting Green's function and self-energy in the device region and can be approximated using standard diagrammatic techniques. Exploiting Wick's theorem a general diagram for the self-energy can be written in terms of bare Green's functions and interaction lines. Any approximation to the self-energy which contains a finite number of such diagrams does, however, violate many conservation laws.

Conserving approximations [5] require the resummation of an infinite number of diagrams and are of paramount importance in nonequilibrium problems as they guarantee satisfaction of fundamental conservation laws such as charge conservation. Examples of conserving approximations are the Hartree-Fock (HF), second Born (2B), GW, T-matrix, and fluctuation exchange (FLEX)

approximations [6]. The success of the GW approximation in describing spectral features of atoms and molecules as well as of interacting model clusters prompted efforts to implement the Meir-Wingreen formula at the GW level in simple molecular junctions and tight-binding models.

The advantage of using molecular devices in future nanoelectronics is not only the miniaturization of integrated circuits. Nanodevices can work at the terahertz regime and hence perform operations in a few picoseconds or even faster. Space and time can both be considerably reduced. Nevertheless, at the subpicosecond time-scale stationary steady-state approaches are inadequate to extract crucial quantities such as, e.g., the switching or charging time of a molecular diode, and consequently to understand how to optimize the device performance.

Recently several practical schemes have been proposed to tackle TD quantum transport problems of noninteracting electrons. In some of these schemes the electronelectron interaction can be included within a TDDFT framework [7] and few calculations on the transient electron dynamics of molecular junctions have been performed at the level of the adiabatic local density approximation. So far, however, no one has extended the diagrammatic MBPT formulation of Meir and Wingreen to the time domain. As in the steady-state case the MBPT formulation allows for including relevant scattering mechanisms via a proper selection of physically meaningful Feynman diagrams. The appealing nature of diagrammatic expansions renders MBPT an attractive alternative to investigate out-of equilibrium systems.

In this paper we consider a time-dependent MBPT formulation of quantum transport which is based on the realtime propagation of the Kadanoff-Baym (KB) equations for open and interacting systems. The KB equations are equations of motion for the nonequilibrium Green's function from which basic properties of the system can be calculated. It is the purpose of this paper to give a detailed account of the theoretical derivation and to extend the numerical analysis to quantum wires connected to two-dimensional leads. For practical calculations we have implemented the fully self-consistent HF, 2B, and GW conserving approximations.

Our results reduce to those of steady-state MBPT implementations in the long-time limit. However, having full access to the transient dynamics we are also able to extract novel information such as the switching and charging times, the time-dependent renormalization of the electronic levels, the role of initial correlations, the time-dependent dipole moments etc.

2. The model of quantum system

We consider a class of quantum correlated open systems (Fig. 1, the left part) (which we call central regions), coupled to noninteracting reservoirs, leads) with the Hamiltonian

$$H(t) = H_C(t) + \sum_{\alpha=L,R} H_\alpha(t) + H_T - \mu N \,, \qquad (1)$$

where H_C, H_α and H_T are the central region, the lead $\alpha = L, R$ and the tunneling Hamiltonians, respectively, and N is the particle number operator coupled to chemical potential μ.

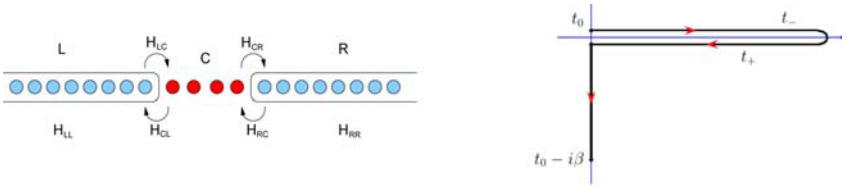

Fig.1. Sketch of the transport setup. The correlated central region (C) is coupled to semi-infinite left (L) and right (R) tight-binding leads via tunneling Hamiltonians **H**_C and **H**C ($\alpha = L, R$).

We assume that there is no direct coupling between the leads. The explicit expressions for these Hamiltonians have the form

$$H_C(t) = \sum_{i,j,\sigma} h_{ij}(t) d^+_{i\sigma} d_{j\sigma} + \sum_{\substack{i,j,k,l \\ \sigma,\sigma'}} v_{ijkl} d^+_{i\sigma} d^+_{i\sigma'} d_{k\sigma'} d_{l\sigma} \ , \tag{2}$$

where i, j label a complete set of one-particle states in the central region, σ, σ' are the spin indices, d^+, d are the creation and annihilation operators, respectively. The one-body part of the Hamiltonian $h_{ij}(t)$ may have an arbitrary time dependence, describing, e.g., a gate voltage or pumping fields. The two-body part accounts for interactions between the electrons, where v_{ijkl} are, for example in the case of a molecule, the standard two-electron integrals of the Coulomb interaction. The lead Hamiltonians have the form

$$H_\alpha(t) = U_\alpha(t) N_\alpha + \sum_{i,j,\sigma} h^\alpha_{ij} c^+_{i\sigma\alpha} c_{j\sigma\alpha} \ , \tag{3}$$

where the creation and annihilation operators for the leads are denoted by c^+ and c. Here $N = \sum_{i\sigma} c^+_{i\sigma\alpha} c_{i\sigma\alpha}$ is the operator describing the number of particles in lead α. The one-body part of the Hamiltonian h_{ij} describes metallic leads and can be calculated using a tight-binding representation, or a real space grid or any other convenient basis set. We are interested in exposing the leads to an external electric field which varies on a time-scale much longer than the typical plasmon time-scale. Then, the coarse-grained time evolution can be performed assuming a perfect instantaneous screening in the leads and the homogeneous time-dependent field $U_\alpha(t)$ can be interpreted as the sum of the external and the screening field, i.e., the applied bias. This effectively means that the leads are treated at a Hartree mean field level. We finally consider the tunneling Hamiltonian H_T,

$$H_T = \sum_{ij,\sigma\alpha} V_{ij\alpha}[d^+_{i\sigma} c_{j\sigma\alpha} + c^+_{j\sigma\alpha} c_{i\sigma}] \ , \tag{4}$$

which describes the coupling of the leads to the interacting central region. This completes the full description of the Hamiltonian of the system. In the next section we study the equations of motion for the corresponding Green's function.

2. Equations for the Keldysh Green functions.

We assume the system to be contacted and in equilibrium at inverse temperature β before time $t = 0$ and described by Hamiltonian H_0. For times $t > t_0$ the system is driven out of equilibrium by an external bias and we aim to study the time-evolution of the electron density, current, etc. In order to describe the electron dynamics in this system we use Keldysh Green's function theory which allows us to include many-body effects in a diagrammatic way. The Keldysh Green's function is defined as the expectation value of the contour-ordered product

$$G_{rs}(z,z') = -i \frac{\mathrm{Sp}\left\{T \exp\left(-i\int dz_1 H(z_1)\right) a_r(z) a_s^+(z')\right\}}{\mathrm{Sp}\left\{T \exp\left(-i\int dz_1 H(z_1)\right)\right\}}, \tag{5}$$

where a^+ and a are either lead or central region operators and the indices *r* and *s* are collective indices for position and spin. The variable z is a time contour variable that specifies the location of the operators on the time contour. The operator T orders the operators along the Keldysh contour displayed in Fig. 2, consisting of two real-time branches and the imaginary track running from τ_0 to $\tau_0 - i\beta$. In the definition of the Green's function the trace is taken with respect to the many-body states of the system.

All time-dependent one-particle properties can be calculated from ***G***. For instance, the time-dependent density matrix is given as

$$n_{rs}(t) = -iG_{rs}(t_-, t_+) \ ,$$

where the times $t\_$ lie on the lower/upper branch of the contour. The equations of motion for the Green's function of the full system can be easily derived from the definition Eq. 5 as

$$\begin{aligned} i\frac{d}{dz} G(z;z') &= 1\delta(z,z') + H(z)G(z,z') + \int_\gamma dz_1 \Sigma^{MB}(z,z_1) G(z_1 z'), \\ -i\frac{d}{dz'} G(z;z') &= 1\delta(z,z') + G(z,z')H(z') + \int_\gamma dz_1 G(z;z_1)\Sigma^{MB}(z_1,z'). \end{aligned} \tag{6}$$

where $\Sigma^{MB}(z,z_1)$ is the many-body self-energy, $H(z)$ is the matrix representation of the one-body part of the full Hamiltonian and the integration is performed over the Keldysh contour (Fig.1, right part). This equation of motion needs to be solved with the boundary conditions

$$G(t_0, z') = -G(t_0 - i\beta, z'),\ G(z, t_0) = -G(z, t_0 - i\beta),$$

which follow directly from the definition of the Green's function Eq. 5. Explicitly, the one-body Hamiltonian H for the case of two leads, left L and right R connected to a central region C, is $H = \| H_{\alpha\beta} \|, (\alpha, \beta = L, C, R)$, where $H_{RL} = H_{LR} = 0$. Here the different block matrices describe the projections of the one-body part H of the Hamiltonian onto different subregions. They are explicitly given as

$$\left(H_{\alpha\alpha}(z)\right)_{i\sigma . j\sigma'} = [h_{ij}(z) + \delta_{ij} + \delta_{ij}\left(U_\alpha(z) - \mu\right)]\delta_{\sigma,\sigma'},$$

$$\left(H_{CC}(z)\right)_{i\sigma . j\sigma'} = [h_{ij}(z) - \delta_{ij}\mu]\delta,\ \left(H_{C\alpha}(z)\right)_{i\sigma . j\sigma'} = V_{ij\alpha}(z)\delta_{\sigma,\sigma'}.$$

The dynamical processes occurring in the central region are described by the Green's function G_{CC} projected onto region C. The many-body self-energy in Eq. 7 has nonvanishing entries only for indices in region C. This implies that $\Sigma^{MB}(G_{CC})$ is a functional of G_{CC} only. From these considerations it follows that in the one-particle basis the matrix structure of Σ^{MB} is given as $\left(\Sigma^{MB}\right)_{\alpha\beta} = \Sigma^{MB}_{CC}(G_{CC})$ $\Sigma^{MB} = \| \Sigma^{MB}_{CC}\delta_{\alpha,C}\delta_{\beta,C} \|$.

The projection of the equation of motion (6) onto regions CC and αC yields

$$\left\{ i\frac{d}{dz} - H_{CC}(z) \right\} G_{CC}(z, z') = \delta(z, z')1 + \sum_\alpha H_{C\alpha} G_{\alpha C}(z, z') + \int_\gamma dz_1 \Sigma^{MB}_{CC}(z, z_1) G_{CC}(z_1 z'), \tag{7}$$

for the central region and

$$\left\{ i\frac{d}{dz} - H_{\alpha\alpha}(z) \right\} G_{\alpha C}(z, z') = H_{\alpha C} G_{CC}(z, z'), \tag{8}$$

for the projection on C. The latter equation can be solved for αC, taking into account the boundary conditions we obtain

$$G_{\alpha C}(z, z') = \int_\gamma dz_1 g_{\alpha\alpha}(z, z_1) H_{\alpha C} G_{CC}(z_1 z'), \tag{9}$$

where the integral is along the Keldysh contour. Here we defined $g_{\alpha\alpha}$ as the solution of

$$\left\{ i\frac{d}{dz} - H_{\alpha\alpha}(z) \right\} G_{\alpha C}(z, z') = H_{\alpha C} G_{CC}(z, z'), \tag{10}$$

with the above mentioned boundary conditions. The function $g_{\alpha\alpha}$ is the Green's function of the isolated and biased α-lead. We wish to stress that a Green's function $g_{\alpha\alpha}$ with boundary conditions automatically ensures the correct boundary conditions for the $G_{\alpha C}(z, z')$ in Eq. (9). Any other boundary conditions would not

only lead to an unphysical transient behavior but also to different steady-state results. This is the case for, e.g., initially uncontacted Hamiltonians in which the equilibrium chemical potential of the leads is replaced by the electrochemical potential, i.e., the sum of the chemical potential and the bias

Taking into account Eq. (9) the second term on the righthand side of Eq. (7) becomes

$$\sum_{\alpha} H_{C\alpha} G_{\alpha C}(z,z') = \int_{\gamma} dz_1 \Sigma_{em}(z,z_1) G_{CC}(z_1 z'), \tag{11}$$

where we have introduced the *embedding* self-energy

$$\Sigma_{\alpha}(z,z') = \sum_{\alpha} \Sigma_{em,\alpha}(z,z') = \sum_{\alpha} H_{C\alpha} g_{\alpha\alpha}(z,z') H_{\alpha C}.$$

which accounts for the tunneling of electrons from the central region to the leads and vice versa. The embedding selfenergies $\Sigma_{em,\alpha}$, are independent of the electronic interactions and hence of G_{CC}, and are therefore completely known once the lead Hamiltonians H_{α} are specified. Inserting Eq. (11) back to Eq. (9) then gives the equation of motion

$$\left\{ i\frac{d}{dz} - H_{CC}(z) \right\} G_{CC}(z,z') = \delta(z,z')1 +$$
$$\sum_{\alpha} H_{C\alpha} G_{\alpha C}(z,z') + \int_{\gamma} dz_1 \left(\Sigma_{CC}^{MB} + \Sigma_{em} \right)(z,z_1) G_{CC}(z_1 z'). \tag{12}$$

Equation (12) is an exact equation for the Green's function G_{CC}. To solve the equation of motion Eq. (12)_ we need to find an approximation for the many-body self-energy $\Sigma_{CC}^{MB}[G_{CC}]$ as a functional of the Green's function G_{CC}. This approximation can be constructed using a variational derivative method which can straightforwardly be extended to the case of contourordered Green's functions. In our case the perturbative expansion is in powers of the two-body interaction and the unperturbed system consists of the noninteracting, but contacted and biased system. We stress, however, that eventually all our expressions are given in terms of fully dressed Green's functions leading to fully self-consistent equations for the Green's function.

2. The equations for a time-dependent current.

An equation for the time-dependent current flowing into the lead α can be derived from the time-derivative of the number of particles in lead α using the equation of motion for the lead's Green's function $G_{\alpha\alpha}$. This yields

$$I_{\alpha}(t) = 2\,\mathrm{Re}\,\mathrm{Sp}\left(G^{<}_{C\alpha}(t,t) H_{\alpha C} \right),$$

where $G^{<}(t,t') = G(t_{\mp}, t'_{\pm})$. If we insert the adjoint of Eq. (9) and extract the different components from the resulting equation, the equation for the cdurrent becomes

$$I_\alpha(t) = 2\,\mathrm{Re}\,\mathrm{Sp}\left(\int_{t_0}^{t} dt_1 G^{<}_{CC}(t,t_1)\Sigma^{A}_{em,\alpha}(t_1,t) + \int_{t_0}^{t} dt_1 G^{R}_{CC}(t,t_1)\Sigma^{<}_{em,\alpha}(t_1,t) - \right.$$

$$\left. - \int_{t_0}^{t} d\tau_1 G^{\urcorner}_{CC}(t,\tau)\Sigma^{\urcorner}_{em,\alpha}(\tau,t) \right).$$

Here $G^{\urcorner}_{CC}(t,\tau) = G(t_{\pm}, t_0 - i\tau)$ and $\Sigma^{\urcorner}_{em,\alpha}(\tau,t) = \Sigma_{em,\alpha}(t_0 - i\tau, t)$. The first two terms in this equation contain integration over early times from t_0 to t and take into account the nontrivial memory effects arising from the time-nonlocality of the embedding self-energy and Green's functions. As anticipated the last term in this equation explicitly accounts for the effects of initial correlations and initial-state dependence. If one assumes that both dependences are washed out in the long-time limit $t \to \infty$, then the last term vanishes and we can safely take the limit $t_0 \to -\infty$.

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МОРФОЛОГИЧЕСКИЕ МОДЕЛИ НАДЕЖНОСТИ ЭНЕРГЕТИЧЕСКОГО ОБОРУДОВАНИЯ

Резюме. Предложена морфологическая модель надежности, основанная на корелляции показателей работоспособности сложных технических объектов с избыточной структурой (резервированием) и структурно (морфологически) подобных им сложных информационных систем. В качестве информационной системы предложено использовать топологически подобные нейронные сети. Описан алгоритм построения и работы с такой моделью.

Отказоустойчивость сложных технических систем закладывается, в