Steady-State Currents through Nanodevices: A Scattering-States Numerical Renormalization-Group Approach to Open Quantum Systems

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We propose a numerical renormalization group (NRG) approach to steady-state currents through nanodevices. A discretization of the scattering-states continuum ensures the correct boundary condition for an open quantum system. We introduce two degenerate Wilson chains for current carrying left- and right-moving electrons reflecting time-reversal symmetry in the absence of a finite bias V. We employ the time-dependent NRG to evolve the known steady-state density operator for a noninteracting junction into the density operator of the fully interacting nanodevice at finite bias. We calculate the differential conductance as function of V, T, and the external magnetic field.

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Introduction.—The description of quantum systems out of equilibrium is one the fundamental challenges in theoretical physics. Even a simple nonequilibrium situation, the current transport through an interacting junction at finite bias is not fully understood. The Coulomb blockade [1] and advent of the experimental realizations of the Kondo effect in such devices [2,3] requires a many-body description at low temperatures.

While the equilibrium dynamics is well understood [4], the nonequilibrium steady-state has been mainly investigated using perturbative approaches [5–8] based on Keldysh theory [9], the Toulouse point [10], and the flow equation [11]. Landauer-Buettiker type approaches [12] treat the charging effect only on a mean-field level by mapping the strongly interacting quantum problem onto a model of noninteracting fictitious particles, unsuitable to describe the Coulomb-blockade physics [1]. In weak coupling and high temperature, the ac and dc transport through molecular wires can be addressed by a quantum master equation for the reduced density matrix of the junction [13]. All those approaches have only a limited validity of their parameter regimes. Recently, Han proposed an alternative perturbative method [14] based on Hershfield's steady-state density operator [15–18]. Based on similar ideas, a scattering-states Bethe-ansatz approach to an interacting spinless quantum dot has been implemented [19] for finite bias.

We present a numerical renormalization group approach [4] to open quantum systems based on scattering states [15]. It combines (i) Wilson chains for single-particle scattering-states proposed below, (ii) Hershfield's steady-state density operator [15] for a noninteracting junctions at finite bias, and (iii) the time-dependent NRG (TD-NRG)[20–22]. Our scattering-states basis will be also useful for Quantum Monte Carlo and density matrix renormalization group (DMRG) approaches [23]. With our nonperturbative method, steady-state currents through interacting nanodevices can be obtained accurately for arbitrary temperatures, magnetic fields, and interaction strength.

Dissipative steady-state currents only occur in open quantum system in which the system size L has been sent to $L \to \infty$ before $t \to \infty$. Transient currents can be calculated on a finite-size tight-binding chain within the TD-NRG as well as the time-dependent DMRG [23,24]. However, such transient currents vanish for $t \to \infty$ or even reverse their sign [24] in those approaches, a consequence of the non-interchangeable limit $t \to \infty$ and $L \to \infty$ [18]. We circumvent this problem by discretizing a single-particle scattering-states basis. Therefore, those states remain current carrying and a faithful representation of an open quantum system.

Theory.—Interacting quantum dots (QD), molecular junctions or other nanodevices are modeled by the interacting region $\mathcal{H}_{\rm imp}$, a set of noninteracting reservoirs $\mathcal{H}_B$, and a coupling between both subsystem $\mathcal{H}_I$: $\mathcal{H} = \mathcal{H}_{\rm imp} + \mathcal{H}_B + \mathcal{H}_I$. Throughout this Letter, we restrict ourselves to a junction with a single spin-degenerate orbital d with energy E_d, subject to an external magnetic field H and an on-site Coulomb repulsion U. The orbital is coupled to a left (L) and a right (R) lead via the tunneling matrix elements $V_{\alpha=L,R}$, and $\mathcal{H}$ given by

$$\mathcal{H} = \sum_{\sigma\alpha=L,R} \int d\epsilon \epsilon c^\dagger_{\epsilon\sigma\alpha} c_{\epsilon\sigma\alpha} + \sum_{\sigma=\pm 1} \left[E_d + \frac{U}{2} - \frac{\sigma}{2} H \right] \hat{n}^d_\sigma + \frac{U}{2} \left(\sum_\sigma \hat{n}^d_\sigma - 1 \right)^2 + \sum_{\alpha\sigma} V_\alpha \int d\epsilon \sqrt{\rho(\epsilon)} \{ d^\dagger_\sigma c_{\epsilon\sigma\alpha} + c^\dagger_{\epsilon\sigma\alpha} d_\sigma \}. \quad (1)$$

Here, $\hat{n}^d_\sigma = d^\dagger_\sigma d_\sigma$, and $c^\dagger_{\epsilon\sigma\alpha}$ creates a conduction electron in the lead α of energy ϵ and density of states $\rho(\epsilon)$.

This Hamiltonian is commonly used to model ultrasmall quantum dots [2,5]. In the absence of the local Coulomb repulsion $\mathcal{H}_U = U(\sum_\sigma \hat{n}^d_\sigma - 1)^2/2$, the single-particle

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problem is diagonalized exactly in the continuum limit [14–17,25,26] by the scattering-states operators

$$\gamma^{\dagger}_{\epsilon\sigma\alpha} = c^{\dagger}_{\epsilon\sigma\alpha} + V_{\alpha}\sqrt{\rho(\epsilon)}G^{r}_{0\sigma}(\epsilon + i\delta) \times \left[d^{\dagger}_{\sigma} + \sum_{\alpha'} \int d\epsilon' \frac{V_{\alpha'}\sqrt{\rho(\epsilon')}}{\epsilon + i\delta - \epsilon'} c^{\dagger}_{\epsilon'\sigma\alpha'} \right] \quad (2)$$

where $\bar{V} = \sqrt{V_L^2 + V_R^2}$, and the Green function $G^{r}_{0\sigma}(z) = [z - (E_d + U/2 - \sigma H/2) - \bar{V}^2 \int d\epsilon \rho(\epsilon)/(z - \epsilon)]^{-1}$. In the limit of infinitely large leads, the single-particle spectrum remains unaltered, and these scattering states diagonalize the Hamiltonian [15] (1) for $U = 0$:

$$\mathcal{H}^{i}_{0} = \mathcal{H}(U = 0) = \sum_{\alpha=L,R;\sigma} \int d\epsilon \epsilon \gamma^{\dagger}_{\epsilon\sigma\alpha}\gamma_{\epsilon\sigma\alpha}. \quad (3)$$

Hershfield has shown that the density operator for such a noninteracting current carrying quantum system retains its Boltzmannian form [15,18] even at finite bias:

$$\hat{\rho}_0 = \frac{e^{-\beta(\mathcal{H}^{i}_{0} - \hat{Y}_0)}}{\mathrm{Tr}[e^{-\beta(\mathcal{H}^{i}_{0} - \hat{Y}_0)}]}, \qquad \hat{Y}_0 = \sum_{\alpha\sigma} \mu_{\alpha} \int d\epsilon \gamma^{\dagger}_{\epsilon\sigma\alpha}\gamma_{\epsilon\sigma\alpha}. \quad (4)$$

The $\hat{Y}_0$ operator accounts for the occupation of the left and right-moving scattering states, and μ_{α} for the different chemical potentials of the leads.

Steady-state NRG.—In order to apply the NRG to such an open quantum systems, the scattering states $\gamma_{\epsilon\alpha\sigma}$ are discretized on a logarithmic energy mesh using the NRG discretization parameter Λ [4]. In contrary to a closed system, however, each of these single-particle states carries a finite current. Even for asymmetric coupling, the spectra of the right and left movers remains symmetric, and the total current vanishes always at zero bias.

Defining the creation operator for a fictitious left- or right-moving $d_{\sigma\alpha}$-orbital $d^{\dagger}_{\sigma\alpha} = \bar{V} \int d\epsilon \sqrt{\rho(\epsilon)}[G^{r}_{0\sigma}(\epsilon + i\delta)]^{*}\gamma^{\dagger}_{\epsilon\sigma\alpha}$, the physical d-level can be decomposed into $d^{\dagger}_{\sigma} = r_R d^{\dagger}_{\sigma R} + r_L d^{\dagger}_{\sigma L}$ by inverting Eq. (2) and using $r_{\alpha} = V_{\alpha}/\bar{V}$. For $U = 0$, the Hamiltonian is diagonal in the left and right movers. We use these $d_{\sigma\alpha}$-orbitals as starting vector of the Householder transformation [4] mapping the discretized scattering-states continuum onto two semi-infinite Wilson chains [4], as depicted in Fig. 1. These chains are almost identical to standard Wilson chain of a noninteracting resonant level model [4]. Each fictitious $d_{\sigma\alpha}$-orbital consists of a normalized linear combination of scattering states $\gamma_{\epsilon\sigma\alpha}$: no auxiliary degrees of freedom has been introduced into the problem.

We divide $G^{r}_{0\sigma}(\epsilon + i\delta)$ into magnitude and phase, $G^{r}_{0\sigma}(\epsilon + i\delta) = e^{i\Phi_{\sigma}(\epsilon)}|G^{r}_{0\sigma}(\epsilon + i\delta)|$, and absorb the energy dependent phase $\Phi_{\sigma}(\epsilon)$ into the scattering-states operators $\gamma_{\epsilon\sigma\alpha}$ by a gauge transformation. Then, the Wilson chains consist only of purely real tight-binding parameters. Diagonalizing the proposed scattering-states Wilson chains yields a faithful representation of the steady-state density operator $\hat{\rho}_0$ for arbitrary bias.

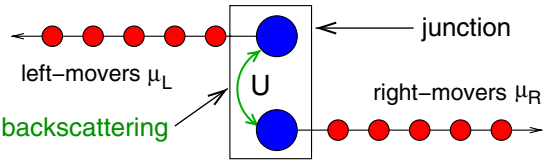

FIG. 1 (color online). The local d-orbital is expanded in left-moving and right-moving scattering states. Each contributions defines one fictitious local orbital $d_{\sigma\alpha}$ of the junction of the scattering-states NRG. The Coulomb repulsion introduces backscattering between left and right movers.

The current operator expanded in scattering states $\gamma_{\epsilon\sigma\alpha}$ acquires an additional energy dependence via the scattering-phase shift $\Phi_{\sigma}(\epsilon)$. In our model (1), however, the current remains connected to the spectral function $A_d(\omega)$ of the retarded nonequilibrium Green function [27]

$$I(V) = \frac{G_0}{e} \sum_{\sigma} \int_{-\infty}^{\infty} d\omega [f(w - \mu_L) - f(w - \mu_R)] \times \pi A_{d\sigma}(\omega)\Gamma \quad (5)$$

in such a scattering-states formulation even for finite U[15,17,28]. $f(\omega)$ denotes the Fermi function, $G_0 = (e^2/h)4\Gamma_L\Gamma_R/(\Gamma_L + \Gamma_R)^2$, $\Gamma_{\alpha} = r_{\alpha}^2 \pi \bar{V}^2 \rho(0)$, $\Gamma = \Gamma_L + \Gamma_R$, and $\pi A_{d\sigma}(\omega) = -\Im m G^{r}_{d\sigma}(\omega + i\delta)$.

Coulomb interaction.—Expanding the operator $\hat{n}^{d}_{\sigma}$ in the orbitals $d_{\sigma\alpha}$ yields two contributions: a density and a backscattering term: $\hat{n}^{d}_{\sigma} = \hat{n}^{0}_{\sigma} + \hat{O}^{\mathrm{back}}_{\sigma}$, with $\hat{n}^{0}_{\sigma} = \sum_{\alpha} r_{\alpha}^2 d^{\dagger}_{\sigma\alpha} d_{\sigma\alpha}$. The backscattering term reads

$$\hat{O}^{\mathrm{back}}_{\sigma} = r_L r_R (d^{\dagger}_{\sigma R} d_{\sigma L} + d^{\dagger}_{\sigma L} d_{\sigma R}) \quad (6)$$

and describes transitions between left and right movers. This term vanishes in the tunnelling regime, where either r_L or r_R vanishes.

We will include the full Coulomb interaction into our theory in two steps. Since H^{0}_{U}, defined as $H^{0}_{U} = \frac{U}{2} \times (\sum_{\sigma} \hat{n}^{0}_{\sigma} - 1)^2$, commutes with $\hat{Y}_0$, the steady-state density operator $\hat{\rho}_0$ evolved into $\tilde{\rho}_0 = \exp[-\beta(\mathcal{H}^{i} - \hat{Y}_0)]/Z$ with $\mathcal{H}^{i} = \mathcal{H}^{i}_{0} + H^{0}_{U}$ proven by the arguments given in Ref. [18]. $\hat{O}^{\mathrm{back}}_{\sigma}$ can be neglected in the tunneling regime where $\hat{\rho} \to \tilde{\rho}_0$. Then, the steady-state spectra is completely determined by a single effective orbital, and the equilibrium spectral function is recovered.

$\mathcal{H}^{i}$ marks the new starting point of our theory. The full Hamiltonian $\mathcal{H}$ of the interacting model differs from $\mathcal{H}^{i}$ by the additional backscattering terms. $\mathcal{H}$ does not commute with $\hat{Y}_0$, and the analytical form of steady-state density operator of the fully interacting problem is not explicitly known [15,18]. We obtain a solution [20–22] by evolving $\tilde{\rho}_0$ with respect to the full Hamiltonian $\mathcal{H}$ into its steady-state value $\hat{\rho}_{\infty} = \lim_{t\to\infty} e^{-i\mathcal{H}t}\tilde{\rho}_0 e^{i\mathcal{H}t}$. In the current-voltage relation (5), the spectral function $A_{d\sigma}(\omega)$

for $U = 0$ is replaced by the nonequilibrium spectral function [28] calculated with respect to $\hat{\rho}_\infty$. The details of this algorithm embedding the calculation of equilibrium spectral functions [29,30] are published in Ref. [22].

Results.—All energies are measured in units of $\Gamma = \pi\bar{V}^2\rho(0)$; a constant band width [4] of $\rho(\omega) = 1/(2D)\Theta(D - |\omega|)$ was used with $D/\Gamma = 10$. The number of retained NRG states was $N_s = 2200$; a $\Lambda = 4$ was chosen. The model lacks channel conservation: only the total charge and z-component of the spin served as quantum numbers. We defined $R = \Gamma_L/\Gamma_R$ and always kept $\Gamma = \Gamma_L + \Gamma_R$ constant. The two chemical potentials μ_α were set to $\mu_L = -r_R^2 V$ and $\mu_R = r_L^2 V$ as function of the external source-drain voltage V consistent with a serial resistor model.

The nonequilibrium spectral function for a symmetric junction is plotted for $U = 8$ and different bias V in Fig. 2(a). Multiple backscattering events cause gain (or lost) of single-particle excitation energy proportional to the applied bias. The Kondo resonance is destroyed with increasing bias due to redistribution of spectral weight towards higher energies. An onset of two weak peaks in the vicinity of the two chemical potentials remains for $|V| > \Gamma$ [16]. For large $R \gg 1$, such backscattering processes are suppressed. The spectral function remains bias-independent. The Kondo resonance remains pinned to $\mu_L \to 0$ as depicted in Fig. 2(b), and we recover the tunneling regime.

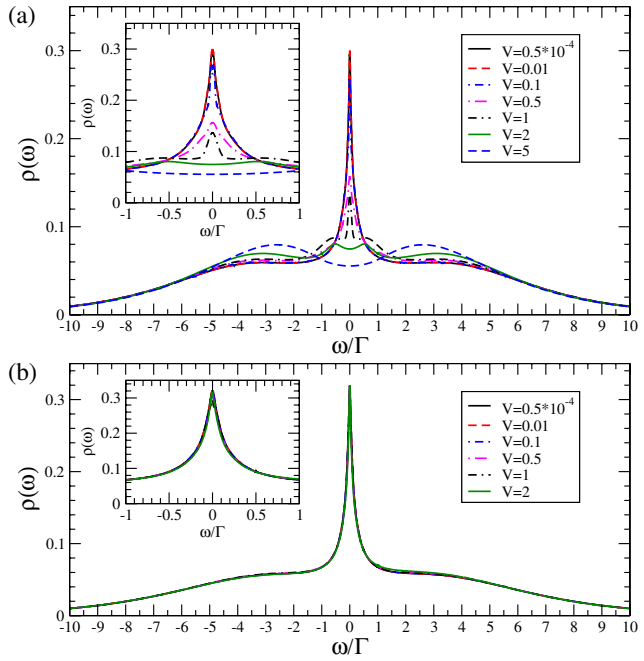

FIG. 2 (color online). Nonequilibrium spectral function for (a) a symmetric junction $R = 1$ at various values of finite bias voltage V, and (b) for a strongly asymmetric junction $R = 1000$. The insets show the evolution of the Kondo-resonance. Parameters: $U = 8$, $\epsilon_f = -4$, and $T \to 0$.

The differential conductance is plotted for different asymmetry ratios R in Fig. 3(a) using the same parameters as in Fig. 2. With increasing R, the nonequilibrium spectral function is less broadened and, therefore, $G(V)$ decreases for large bias voltage. Asymptotically, G approaches the equilibrium t-matrix which is the exact result for $R \to \infty$ and $T \to 0$.

The effect of an external magnetic field onto the differential conductance is shown in Fig. 3(b). An increasing magnetic field splits the zero-bias anomaly which is further suppressed by the finite bias in a symmetric junction. This field dependence has been used in experiments [2] as hallmark for the Kondo physics at low temperatures.

In Fig. 3(c), the NRG conductance for $U = 5$ is compared to the result of Ref. [16]. Both curves agree for low bias. The NRG result shows a weaker decay of the zero-bias anomaly with increasing bias with a less pronounce maximum at large bias. The symmetrized equilibrium t-matrix [4] is added for comparison as a dashed line.

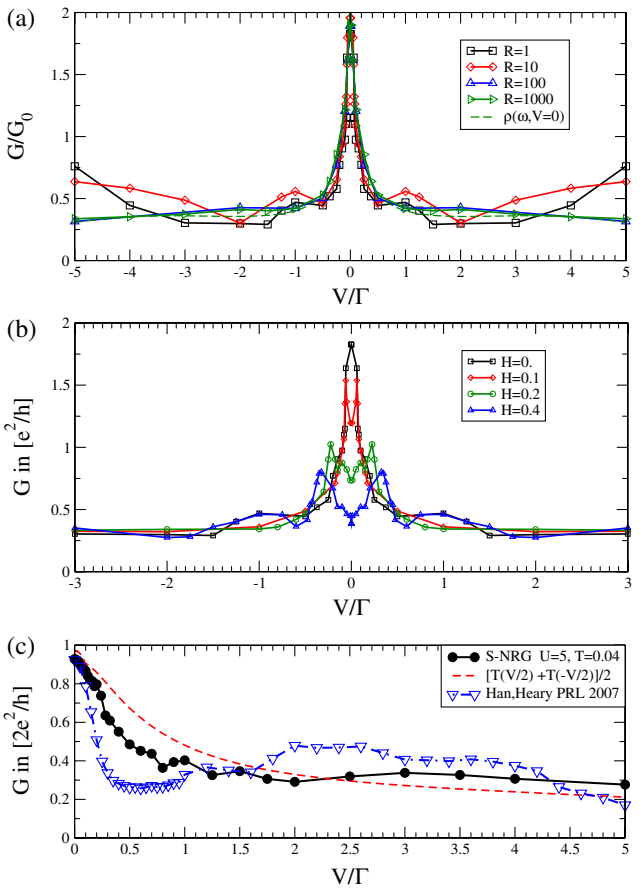

FIG. 3 (color online). The differential conductance $G = dI/dV$ as function of the bias voltage (a) for different asymmetry factors R, (b) for different magnetic field $H = 0$, 0.1, 0.2, 0.4, and $R = 1$. Parameters: as in Fig. 2. (c) Comparison between the results for $U = 5$ from Ref. [16] and the NRG calculation at $T/\Gamma = 0.04$ and $R = 1$ using z-averaging over 4 z-values [20,21].

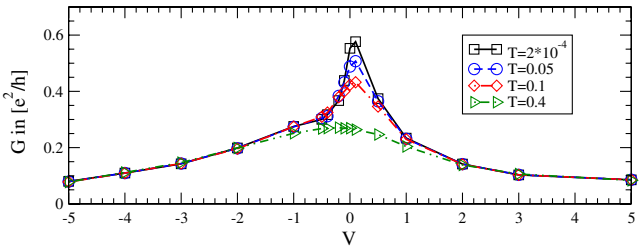

FIG. 4 (color online). The differential conductance G as function of the bias voltage for different temperatures. Parameters $R = 10$, $\epsilon_f = -1.5$, and $U = 12$.

The more generic case of an asymmetric junction with respect with a relatively large local Coulomb repulsion is plotted in Fig. 4. The differential conduction reflects the lack of symmetry under source-drain voltage reversal. As depicted, the zero-bias peak vanishes with increasing temperature.

Conclusion.—A powerful new approach to the steady-state currents through nanodevices has been presented. We have introduced a NRG method based on scattering states to incorporate the correct steady-state boundary condition of current carrying systems. The steady-state density operator [15] of a noninteracting junction is evolved into the one of the interacting nanodevice using the TD-NRG [20]. We have established an accurate solution for the strong-coupling regime and calculated steady-state currents for arbitrary ratios R at finite bias. The tunneling regime is included as an exact limit. Our approach does not suffer from any current reflection inherent to numerical simulations of closed quantum systems [24]. We have concentrated on the low-temperature properties of the nanodevice since the combination of arbitrary bias, large Coulomb repulsion, and finite magnetic field remains the most difficult regime for all perturbative methods. However, the NRG is equally suitable to calculate the crossover from the low to the high-temperature regime as demonstrated in Fig. 4. An experimental hallmark [2] for Kondo physics, the splitting of the zero-bias Kondo peak with increasing magnetic field, is correctly described by our approach for arbitrary temperature, bias, and field strength.

This theory can be extended to more complicated multi-orbital models. Equation (5) must be modified and requires more complex correlation functions. Since single-particle scattering states can always be obtained exactly, the construction of the Wilson chain parameters is straight forward using the corresponding expansion of the local degrees of freedom and combining it with the transformation used for nonconstant density of states [4].

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