Non-equilibrium quantum dynamics of the magnetic Anderson model

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Abstract. We study the non-equilibrium dynamics of a spinful single-orbital quantum dot with an incorporated quantum mechanical spin-1/2 magnetic impurity. Due to the spin degeneracy, double occupancy is allowed, and Coulomb interaction together with the exchange coupling of the magnetic impurity influence the dynamics. By extending the iterative summation of real-time path integrals (ISPI) to this coupled system, we monitor the time-dependent non-equilibrium current and the impurity spin polarization to determine features of the time-dependent non-equilibrium dynamics. We particularly focus on the deep quantum regime, where all time and energy scales are of the same order of magnitude and no small parameter is available. We observe a significant influence of the non-equilibrium decay of the impurity spin polarization both in the presence and in the absence of Coulomb interaction. The exponential relaxation is faster for larger bias voltages, electron–impurity interactions and temperatures. We show that the exact relaxation rate deviates from the corresponding perturbative result. In addition, we study in detail the impurity’s back action on the charge current and find a reduction of the stationary current for increasing coupling to the impurity. Moreover, our approach allows us to systematically distinguish mean-field Coulomb and impurity effects from the influence of quantum fluctuations and flip-flop scattering, respectively. In fact, we find a local maximum of the current for a finite Coulomb interaction due to the presence of the impurity.
1. Introduction

Diluted magnetic semiconductors [1–4] are an important class of materials for the spintronics community since they combine the properties of (ferro-)magnets and semiconductors. Thus they allow for the all-electrical control of the magnetic degrees of freedom of spintronic devices. The magnetization of semiconducting devices is mainly caused by the interaction of magnetic impurities in the sample with itinerant charge carriers. To understand the microscopic details of these magnetic materials, reduced model systems are required in which the individual constituents are well under control. This allows us to study fundamental questions concerning the interplay of coherent quantum dynamics and dissipation under non-equilibrium conditions. An ideal candidate for such a model system is a small quantum dot (QD) that connects metallic leads and also carries a magnetic degree of freedom described by a quantum mechanical spin (magnetic Anderson model).

Magnetic QDs have been studied experimentally in ensembles that are particularly suited to investigation by laser and electromagnetic fields [5–11]. They are designed with standard lithographic methods and are technologically well established. Moreover, embedding individual Mn ions into QDs and studying the electrical properties is possible [12–14]. Small QDs with very few charge carriers and a single magnetic impurity may become important candidates for efficient high-density spintronic devices.

Although Mn ions in QDs usually have spin larger than $S = 1/2$, there are good reasons to study the low-spin case first. From a methodological point of view, this simple model is an ideal basis on which to develop the numerical tools necessary for treating the real-time dynamics of more general coupled electron–impurity spin systems as well. In addition, the magnetic Anderson model serves as a generic model to study electronic transport through...
magnetic atoms or molecules placed between the tip of a scanning tunnelling microscope and a substrate \[15–17\]. Likewise, our model is a phenomenological basis for microscopic studies of molecular junctions based on organic radicals \[18\]. We mention the switching of the spin state of the central iron ion in a single molecular complex in a double layer on gold by a low-temperature scanning tunnelling microscope \[19\] for which our model is also applicable. Finally, it also mimics the features of the dynamics of electrons in QDs that are subject to hyperfine interactions with the nuclei of the host material. In this respect, an understanding of the electron–impurity coupling and its influence on the dephasing and relaxation times of qubits are essential for the experiments realized in \[20, 21\].

Non-equilibrium quantum transport in strongly correlated systems remains a challenging problem. In particular, reliable theoretical treatments prove to be difficult when no small parameter is present in the system, i.e. in the deep quantum regime. Approximate methods are often based on advanced perturbative treatments such as quantum master equations \[22–24\], which are based on Markovian approximations and weak tunnelling coupling. The renormalization group (RG) approach \[25–30\] as well as the functional RG method are able to capture essential non-equilibrium features \[31–34\]. Real-time density matrix RGs \[35–38\] require one to represent the system plus the macroscopic reservoirs by a large but finite lattice. Due to the possible appearance of finite size effects, the maximal propagation times are limited.

Quantum Monte-Carlo (QMC) concepts are a priori numerically exact and have been adopted for non-equilibrium quantum transport \[39–43\]. As opposed to the fermionic sign problem, which shows up in equilibrium simulations of quantum many-body systems, the real-time (non-equilibrium) QMC weight function is itself highly oscillatory and causes the dynamical sign problem. Reaching the stationary non-equilibrium state at asymptotic times remains notoriously difficult \[39–44\]. An analytic continuation of the ‘complex voltage plane’ via imaginary ‘Matsubara voltages’ tries to circumvent the dynamical sign problem. However, the postprocessed back transformation to real times is plagued by numerical instabilities \[45, 46\]. Recently, long-time and steady state results were obtained by a QMC sampling of the diagrammatic corrections to the non-crossing approximation \[47\], stating the reduction of the sign problem.

In this work, we adopt the method of the iterative summation of path integrals (ISPI); see \[48\] for the magnetic Anderson model. This approach evaluates a real-time path integral expression for the Keldysh partition function in a numerically exact manner and is particularly suitable for nonlinear transport. Recently, this scheme was carefully verified by a comparison to existing approximations in the appropriate parameter regimes for the single-impurity Anderson model \[48\]. Furthermore, the prediction of a sustained Franck–Condon blockade in the deep quantum regime has been reported based for ISPI data as well \[49\]. In contrast to the stochastic evaluation of the real-time path integral in the QMC approach, the ISPI scheme calculates the real-time path integral deterministically. Hence the sign problem is avoided. The fact that metallic leads at finite temperature suppress all electronic correlations exponentially beyond a finite memory time window is exploited by the ISPI method. This allows for an iterative propagation of an augmented reduced density operator of the system to arbitrary long times. By construction, the technique is limited to finite temperatures and/or finite bias voltages and not too large system sizes. Whenever numerical results are converged with respect to the memory time, they are numerically exact. Recently, Segal et al \[50\] provided an alternative formulation of this approach in terms of Feynman–Vernon-like influence functionals.
We theoretically investigate the real-time dynamics of a single-level quantum dot (SLQD) containing a magnetic impurity with spin-1/2. Exchange interaction with on-dot electrons results in dissipation, induced by the metallic leads to the localized impurity in the QD. The deep quantum regime, characterized by the absence of a small parameter, is the focus of the present paper, i.e. all interaction strengths are of the same order of magnitude. In particular, we are interested in the nonlinear transport regime, where a finite bias voltage is applied and linear response theory is no longer applicable. The real-time relaxation of the impurity spin as a function of various system parameters is investigated. Due to the additional degree of freedom of the magnetic impurity, an extension of the ISPI scheme is necessary. Although the inclusion of a magnetic impurity affects the single-particle dynamics in the first place, coupling of the impurity to the electronic density on the dot renders this extension non-trivial. Our accurate results show that in the considered crossover regime, the non-equilibrium charge current significantly influences the quantum relaxation dynamics of the impurity. Likewise, the back action of the impurity dynamics on the non-equilibrium current becomes significant. Most importantly, we clearly show that this crossover regime is not accessible by a perturbative means.

The structure of the paper is as follows. After introducing the model in section 2, we express the Keldysh partition function as a real-time path integral in section 3 and show how to evaluate this path sum by a deterministic iteration scheme. We calculate in section 4 the stationary transport current and the impurity spin dynamics. Some of the results are compared with the outcomes of perturbative methods for appropriate parameter combinations. We analyse the influence of the non-equilibrium current on impurity relaxation. Furthermore, we present the results on the stationary tunnelling current in the deep quantum regime, where the rate equation results are not reliable. The dependence of the relaxation rate on various model parameters is presented.

2. The model system

We extend the single-impurity Anderson model to the electronic degree of freedom of a QD coupled to two metallic leads (L/R) via tunnelling barriers in order to study magnetic QDs, see figure 1 for a sketch. We assume equal tunnelling barriers on the left and right sides; the generalization to the asymmetric case is straightforward. The total Hamiltonian is given by

$$H = H_{\text{dot}} + H_{\text{leads}} + H_T,$$

where we use units such that $\hbar = 1$. The Hamiltonian of the magnetic dot

$$H_{\text{dot}} = H_{\text{el, dot}} + H_{\text{imp}} + H_{\text{int}}$$

includes the electronic part $H_{\text{el, dot}}$ and the part $H_{\text{imp}}$ for describing the spatially fixed magnetic impurity as well as the coupling term between the electron and the impurity denoted as $H_{\text{int}}$.

We write the electronic part of the QD as

$$H_{\text{el, dot}} = H_{\text{el, dot}}^0 + H_{\text{el, dot}}^U = \sum_\sigma \epsilon_\sigma d_\sigma^\dagger d_\sigma + Ud_\uparrow^\dagger d_\downarrow^\dagger d_\downarrow,$$

where the operator $d_\sigma$ annihilates a dot electron with spin $\sigma$. The dot is formed by a single spin degenerate level, which can be controlled by a gate electrode that shifts the electrostatic potential of the dot $\Phi_D$. Hence, the electronic degree of freedom can assume four values $\{0, \uparrow, \downarrow, d\}$, indicating whether the dot is empty (0), contains one electron with spin $\sigma = \uparrow, \downarrow = \pm 1$ and

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Figure 1. (a) Two metallic leads are coupled to a QD via tunnelling barriers. The gate electrode allows us to tune the electrostatic potential $\Phi_D$, and the bias voltage $V$ induces an electron current. The QD has a single orbital electronic level that is empty or is occupied either by one electron in a spin up or down state (b) or by two electrons in a Coulomb-interacting singlet state (c). Moreover, the QD carries a localized magnetic impurity $M$ with spin 1/2. In panel (b), a single dot electron interacts with $M$ via an exchange interaction $J$, while in panel (c) only Coulomb interaction is present for double occupation.

Energy $\epsilon_\sigma = \Phi_D + \sigma \Delta/2$ or is in a spin singlet state with double occupation (d). The Coulomb repulsion is modelled via $U > 0$ when the dot is in the doubly occupied state $d$. The Zeeman level splitting $\Delta$ might be present due to possible external or internal crystallographic magnetic fields.

The magnetic impurity (quantum spin) is included via the Hamiltonian

$$H_{\text{imp}} = \frac{\Delta_{\text{imp}}}{2} \tau_z,$$

with the Zeeman energy $\Delta_{\text{imp}}$. Spin operators of the impurity are given in terms of the Pauli matrices $\tau_{x,y,z}$ with $\tau_\pm = \tau_x \pm i \tau_y$. The impurity spin $\tau$ and the dot electron spins $\sigma$ are coupled by the exchange interaction of strength $J$, which is captured by the Hamiltonian

$$H_{\text{int}} = J \tau_z (d_\uparrow \sigma d_\uparrow - d_\downarrow \sigma d_\downarrow) + \frac{J}{2} (\tau_+ d_\uparrow d_\downarrow + \tau_- d_\downarrow d_\uparrow).$$

While the first term shifts the single-particle energies, the second term induces mutual flips of an electron and the impurity spin, which we denote as flip-flop processes. The interaction vanishes for double occupation of the dot; then two electrons form a spin-singlet state.

As usual, we model the non-interacting leads by $H_{\text{leads}} = \sum_{k \sigma} \epsilon_k c_k^\dagger c_\sigma$, where $c_{k\sigma}$ annihilates an electron with spin $\sigma$ and wave vector $k$ in lead $p = L, R = \pm 1$. A bias voltage $V$ is symmetrically applied between the (thermal) leads and shifts their electrochemical potentials such that $\mu_p = p eV/2$. Finally, the tunnelling Hamiltonian $H_T = \sum_{k \sigma} \gamma d_\sigma^\dagger c_{k\sigma} + \gamma^* c_{k\sigma}^\dagger d_\sigma$ describes the energy- and spin-independent tunnelling of electrons with the amplitude $\gamma$. We assume a constant density of states $\rho(\epsilon_F)$ around the Fermi energy and work in the wide-band limit. Then, the tunnelling is parameterized by the parameter $\Gamma' := 2\pi |\gamma|^2 \rho(\epsilon_F)$.
3. The iterative summation of real-time path integrals approach

In order to determine the non-equilibrium electron current and to study the relaxation properties of the impurity, we generalize the approach of ISPI of Weiss et al [48]. This is a non-trivial step and requires us to incorporate the impurity quantum dynamics consistently within the quantum many-body formalism.

3.1. Path integral, generating function and short-time propagator

The ISPI approach is based on the Keldysh partition function \( Z = \text{Tr}[U_K[H] \rho(-\infty)] = 1 \), where \( U_K[H] = T_K \exp(-i \int_K H dt) \) is the propagator along the Keldysh contour \( K \). The time ordering operator is \( T_K \) and \( \rho(-\infty) \) is the density matrix of the system’s initial state [51, 52]. A generating function \( Z[\eta(t)] \) is constructed to calculate the expectation value of an operator \( O(t) \) via

\[
\langle O \rangle_t = \left. \frac{\delta \ln Z[\eta(t)]}{\delta \eta(t)} \right|_{\eta=0},
\]

with \( Z[\eta(t)] = \text{Tr}[U_K[H + i\eta(t) O] \rho(-\infty)] \). Then, \( Z[\eta(t)] \) is represented by a (fermionic) path integral. Throughout this work, we assume an initially \(( t_i = -\infty)\) empty QD prepared in the spin-up impurity state. The full Keldysh time interval \( \Delta_t := t_f - t_i = N\delta_t \) is discretized into time steps \( \delta_t \). A short-time propagator \( U_{\delta_t} \) is introduced such that it is related to the exact propagator \( U(t + \delta_t, t) = U_{\delta_t} + O(\delta_t^2) \). Subsequently, a complete basis of fermionic coherent states is inserted between every two short-time propagators, accounting for the Coulomb and the flip-flop interactions.

To construct a particular \( U_{\delta_t} \), we separate the total Hamiltonian \( H \) into the diagonal part \( H_0 \) and a non-diagonal part \( H_1 \) with respect to the appearing dot operators \( d_\sigma \) and \( d_\sigma^\dagger \). This yields \( H = H_0 + H_1 \) with

\[
H_0 = H_{\text{dot}}^0 + H_{\text{imp}} + H_{\text{int}}^\dagger + H_{\text{leads}},
\]

\[
H_1 = H_{\text{dot}}^U + H_{\text{int}}^\perp + H_{\text{I}}.
\]

In the interaction picture, the full real-time propagator from the initial to the final time can be written as

\[
U(t_f, t_i) = \sum_{N=2}^\infty (-i)^{N-2} \int_{t_i}^{t_f} \cdots \int_{t_{N-2}}^{t_i} dt_2 dt_3 \cdots dt_{N-1} \times U_0(t_f, t_{N-1}) H_1 \cdots H_1 U_0(t_3, t_2) H_1 U_0(t_2, t_1),
\]

where the Keldysh contour is divided into \( N - 1 \) pieces of free propagation by \( U_0(t_{k+1}, t_k) \) that are connected by \( N - 2 \) interaction vertices \(-iH_{I} dt_k\) acting during the transition time \( dt_k \). Here, we define \( t_1 := t_i \) and \( t_N := t_f \). When \( t_{k+1} = t_k + \delta_t \) and in the limit of very small \( \delta_t \), the system can either propagate freely during \( \delta_t \) via the free propagator \( U_0(\delta_t) = U_0(t_{k+1}, t_k) = U_0(t_{k+1} - t_k) \) or undergo at most one transition induced by \( H_1 \) within the interval \( 0 < t' < \delta_t \). Hence, the short-time propagator takes the form

\[
U_{\delta_t} = U_0(\delta_t) - i \int_0^{\delta_t} dt' U_0(\delta_t - t') H_1 U_0(t') = :U_0(\delta_t) (\mathbb{1} - iH_1 \delta_t) + O(\delta_t^2),
\]
where the commutator $[U_0(t), H_1] = O(t)$ is used to bring $H_1$ to the right of $U_0(t)$. The remaining time integral is evaluated exactly up to order $O(\delta^2 t^3)$. A comparable error is obtained by normal ordering, denoted by colons in equation (8).

### 3.2. Discrete Hubbard–Stratonovich transformation

Next, we treat the Coulomb term $H_{\text{dot}}^U$ in a similar way as in [48]. Since $[H_0, H_{\text{dot}}^U] = 0$, the propagator for the system with Coulomb interaction factorizes into a free part $U_0(\delta_t)$ and the interacting part $\exp\{-i H_{\text{dot}}^U \delta_t\}$. We apply the Hubbard–Stratonovich (HS) transformation and obtain

$$
\exp\{-i H_{\text{dot}}^U \delta_t\} = \frac{1}{2} \sum_{\zeta = \pm 1} \exp\left\{-i \left[ \frac{U}{2} (n_\uparrow + n_\downarrow) + i \zeta \lambda_{\delta_t} (n_\uparrow - n_\downarrow) \right] \delta_t \right\},
$$

with the HS parameter $\lambda_{\delta_t}$ defined via

$$
\lambda_{\delta_t} \delta_t = \sinh^{-1} \sqrt{\sin[U \delta_t / 2]} + i \sin^{-1} \sqrt{\sin[U \delta_t / 2]}.
$$

The variable $\zeta = \pm 1$ is interpreted as a fluctuating Ising-like spin field. Note that the solution given in equation (10) is uniquely defined as long as $0 \leq U \delta_t \leq \pi$. By this step, the interacting problem with local-in-time Coulomb repulsions is effectively mapped to a non-interacting problem, at the price that the appearing spin fields interact over a finite time range with each other. In general, the condition $\delta_t \Gamma \ll 1$ is needed to minimize the systematic error of order $\delta^2 t^3$. If $\delta_t$ is bounded from below, however, $U$ should be adjusted in agreement with the condition in equation (10). The exponential in equation (9) commutes with $H_0$ and we can write the full short-time propagator as

$$
\exp\{-i (H_0 + H_{\text{dot}}^U) \delta_t\} = \frac{1}{2} \sum_{\zeta = \pm 1} \exp\{-i H_0^\zeta \delta_t\},
$$

thereby absorbing the classical part of the Coulomb interaction into the single-particle dot energies according to

$$
\epsilon_\sigma^\zeta(\delta_t) = \epsilon_\sigma + \frac{U}{2} + i \sigma \zeta \lambda_{\delta_t}.
$$

In passing, we note that due to the imaginary energy component, $H_0^\zeta$ should not be considered as a Hamiltonian. Instead, we obtain a short-time propagator by enforcing normal ordering, i.e.

$$
U_{\delta_t}^U := \frac{1}{2} \sum_{\zeta = \pm 1} \exp\{-i H_0^\zeta \delta_t\}.
$$

Combining the short-time propagators again into the full path integral, the path sum extends over all tuples $\{\zeta\} := (\zeta_N, \ldots, \zeta_1)$ of the HS fields.

### 3.3. The remaining interaction terms

The tunnelling term $H_T$ is quadratic in the number of fermions but contains both dot and lead operators. Therefore, the stationary state of the isolated system is, in general, not an eigenstate of the system with tunnelling. However, for arbitrary electronic coherent and impurity states
\[ |\Psi^\tau\rangle \equiv |\Psi\rangle|\tau\rangle, \] the matrix elements can be written as

\[ \langle \Psi^\tau | : \exp\{-iH_0^\tau \delta_i\} (\mathbb{1} - iH_T \delta_i) : |\Psi^\tau\rangle = \langle \Psi^\tau | : \exp\{-i(H_0^\tau + H_T) \delta_i\} : |\Psi^\tau\rangle + O(\delta_i^2). \]  

Hence, the short-time propagator is obtained by adding the coupling term \( H_T \) to \( H_0^\tau \) in equation (12). To be specific, we introduce the total electronic coherent state as

\[ |\Psi\rangle \equiv \prod_{k\sigma} (1 - \zeta_{k\sigma p} c_{k\sigma p}^\dagger) \prod_\sigma (1 - \delta_\sigma d_\sigma^\dagger) |0\rangle, \tag{14} \]

where \( \delta_\sigma \) and \( \zeta_{k\sigma p} \) are Grassmann numbers for dot and lead electrons.

Flip-flops of the electron and impurity spin are mediated by the non-diagonal exchange coupling term \( H_{\text{int}}^\tau \) in equation (6). According to equation (8) it enters the short-time propagator as

\[ U_{\delta_t} = \frac{1}{2} \sum_{\zeta = \pm 1} : \exp\{-i(H_0^\tau + H_T) \delta_i\} (\mathbb{1} - iH_{\text{int}}^\perp \delta_i) :. \tag{15} \]

In fact, this structure of the short-time propagator motivates our choice of a ‘mixed’ basis of electron coherent states \( |\Psi\rangle \) and impurity states \( |\tau\rangle \). Most importantly, a straightforward derivation of a path integral in this basis becomes possible as the paths are separated into parts that contribute to the matrix element \( \langle \Psi^\tau | U_{\delta_t} | \Psi^\prime\rangle \) for either aligned (\( \propto \mathbb{1} \)) or opposite (\( \propto H_{\text{int}}^\perp \)) impurity spin orientations. This form is particularly useful with respect to a numerical summation over discrete impurity paths. An equivalent short-time propagator in the form of a single exponential \( : \exp\{-iH^\tau \delta_i\} : \) with \( H^\tau := H_0^\tau + H_T + H_{\text{int}}^\perp \) is much more cumbersome.

In contrast, the short-time propagator \( \mathbb{1} - iH^\tau \delta_i \), despite being exact up to \( O(\delta_i^2) \), is not convenient for constructing a path integral. Consider the case of opposite impurity spin states \( \tau \neq \tau' \). The phase accumulated by the free propagation between two instantaneous flip-flop events is missing, i.e.

\[ \langle \Psi^\tau | \mathbb{1} - iH^\tau \delta_i | \Psi^\tau\rangle_{\tau \neq \tau'} = -\frac{iJ_1}{2} (\delta_{\tau', \tau + 1} \delta_{\tau} + \delta_{\tau', \tau - 1} \delta_{\tau}) e^{\Psi^\tau}. \tag{16} \]

Such a propagator would be correct only if the transition process lasts the entire time span \( \delta_i \) instead of being instantaneous. In the resulting path integral, the system does not propagate freely between consecutive flip-flop processes with the resulting continuous limit \( \delta_i \to 0 \) being unphysical.

3.4. Constructing the full path integral

The path integral for the generating function \( \mathcal{Z}[\eta] \) is obtained by using equation (15) and the corresponding Grassmann fields \( \Psi \) as well as the discrete paths \( \{\tau\} \) and \( \{\zeta\} \). This yields

\[ \mathcal{Z}[\eta] = \sum_{\{\tau, \zeta\}} \int D[\Psi] (-1)^\ell \left(-\frac{iJ_1}{2}\right)^m P[\{\tau\}] e^{i\mathcal{L}[\Psi, \psi, \tau, \zeta]}, \tag{17} \]

where the path sums over the impurity and HS spin fields are performed over the \( 2N \) tuples \( \{\tau_j\} = (\tau_{2N}, \ldots, \tau_1) \) and \( \{\zeta_j\} = (\zeta_{2N}, \ldots, \zeta_1) \) with \( \tau_j, \zeta_j = \pm 1 \). Within an impurity path \( \{\tau\} \), \( m \) flip-flop transitions occur on the Keldysh contour, where \( \ell \) of them lie in the lower branch. In the following, we discuss the building blocks of equation (17).
Figure 2. (a) Exemplary impurity path (blue line) for which the flip-flop polynomial is constructed. The Keldysh contour is divided into $N-1=8$ segments of length $\delta_t$ between $2N=18$ time vertices. The impurity path (tuple of black and red arrows) realizes $m=8$ flip-flops with five flip-flops on the forward and $\ell=3$ flip-flops on the backward branch. First, the flip index tuples $T_{\pm}^{\text{flip}}$ are constructed by assigning to each flip-flop the index of the Keldysh time that is later with respect to the real time. Hence, if two consecutive spins have opposite orientations, the corresponding flip-flop has the time index of the spin on the right-hand side of the flip (marked in red). We have $T_{\text{flip}}^{+}=(8, 6, 5, 4, 2)$ and $T_{\text{flip}}^{-}=(14, 12, 11)$. The polynomial written in panel (b) follows upon using equation (22). We note that the electron’s spin flips in the opposite direction as compared to the impurity (not shown).

In equation (17), the action $S = S_{\text{imp}} + S_{\text{el}} + S_{O}[\eta]$ is given by the sum of the free impurity action and the electronic action. Due to possible source terms, depending on the observable of interest $O(t)$, $S_{O}[\eta]$ is added when necessary (see below). The electronic action contains all the coupling terms of the electrons to the impurity, to the leads (via the tunnelling) and to the HS fields. In particular, we have the action of the free impurity

$$S_{\text{imp}} = -\frac{\Delta_{\text{imp}} \delta_t}{2} \sum_{k=2}^{N} (\tau_k - \tau_{2N-k+1}) = -\frac{\Delta_{\text{imp}}}{2} \int_{K} dt \tau(t), \quad (18)$$

with the discrete and continuous representation given as the first and the second expression, respectively. This equation also illustrates how a well-defined continuous limit of a discrete path can be obtained (cf figure 2). The electronic action $S_{\text{el}}$ can be represented as

$$S_{\text{el}} = S_{\text{el}}^{\dot{\Psi}} + S_{\text{leads}} + S_{T} = \int_{K} dt \int_{t'} dt' \bar{\Psi}(t)(G^{\text{el}})^{-1}_{t,t'}\Psi(t'), \quad (19)$$
with the inverse electronic Keldysh Green’s function \( (G^{el})^{-1} \) naturally given in terms of the action \( S_{el} \) with the three contributions

\[
S_{el_{\text{dot}}} = \sum_\sigma \int_{\mathcal{K}} dt \delta_{\sigma}(t) [i \partial_t - E_\sigma(t)] \sigma_\sigma(t),
\]

\[
S_{\text{leads}} = \sum_{k \sigma p} \int_{\mathcal{K}} dt \delta_{k \sigma p}(t) [i \partial_t - \epsilon_k] \delta_{k \sigma p}(t),
\]

\[
S_T = \sum_{k \sigma p} \int_{\mathcal{K}} dt [\gamma \delta_{\sigma}(t) \delta_{k \sigma p}(t) + \gamma^* \delta_{k \sigma p}(t) \partial_\sigma(t)].
\]

We note that one of the time integrations in equation (19) is trivial, since \( (G^{el})^{-1} \) is proportional to \( (t - t') \). Since the bias voltage enters through the respective lead equilibrium density matrix, see equation (23), electronic energies are the same in both the leads. We have used the effective dot energy in equation (20) defined as

\[
E_\sigma(t) = E_\sigma^*(t) \equiv \epsilon_\sigma + \frac{U}{2} + J \sigma \tau(t) + i \sigma \zeta(t) \lambda_{\delta_i},
\]

on the forward branch, while \( E_\sigma^*(t) = E_\sigma^*(t)^* \) on the backward branch.

The polynomial \( P[\{\tau\}] \) in equation (17) depends on the impurity path \( \{\tau\} = \{\tau^+\} ((\tau^-)) \) for the forward (backward) branch of the contour. Then, we collect all indices of the flips into the tuple \( T^+_\text{flip} = (k_{m-1}, \ldots, k_1) \) (sorted in ascending order) along the forward path \( \{\tau^+\} := (\tau_N, \ldots, \tau_1) \) with \( \tau_k \neq \tau_{k+1} \) for all \( k^+ \in T^+_\text{flip} \). Accordingly, \( T^-_\text{flip} = (k_{N+1}, \ldots, k_N) \) is the tuple of ascending flip indices along the backward path \( \{\tau^-\} := (\tau_{2N}, \ldots, \tau_{N+1}) \) with \( \tau_k \neq \tau_{k+1} \) for all \( k^- \in T^-_\text{flip} \). Note that a flip index on the backward path is labelled according to the smaller step index of the flipping spins corresponding to the later time. The impurity polynomial can be expressed in terms of the Grassmann fields as

\[
P[\{\tau\}] := \prod_{j \in T^+_\text{flip}} \tilde{\delta}^{+1}_{t_j} \tilde{\sigma}^{-1}_{t_j} \prod_{k \in T^-_\text{flip}} \tilde{\delta}^{+1}_{t_k} \tilde{\sigma}^{-1}_{t_k}.
\]

Figure 2 illustrates an example of an impurity path. The fields of the forward and backward parts of the continuous inverse Green’s function \( (G^{el})^{-1} \) are not coupled since the corresponding Hamiltonian is diagonal. However, the associated discrete version connects fields from both branches via the upper right element \((1, 2N)\), which is due to the system’s initial state \( \rho(t_i) \); for details see [52]. Throughout this work, we assume factorizing initial conditions

\[
\rho(t_i) = |0, \tau_1\rangle \langle 0, \tau_1| \rho_L \rho_R,
\]

where \( \rho_p \propto \exp(-\beta \sum c_{k p} (\epsilon_k - \mu_p) c^\dagger_{k p} c_{k p}) \) is the equilibrium density matrix of the lead \( p = L/R \) at a temperature \( T \) with \( \beta = (k_B T)^{-1} \), and \( |0, \tau_i\rangle \) denotes the empty dot with the impurity in the initially prepared orientation \( \tau_i = | \uparrow \rangle \).

When constructing the action \( S_{el}[\eta] \) for an observable \( O(t_m) \), evaluated at the measurement time \( t_m \), we replace every instance of an impurity or electron operator in the observable by the corresponding spin- and Grassmann fields from the forward branch at the time step closest to \( t_m \). This choice is arbitrary and a replacement on the backward branch would not change physical results. Let us assume that \( t_m - t_i = (k_m - 1) \delta_i \). The electron operators are replaced by their Grassmann field with time index \( k_m \). Correspondingly, we substitute the Pauli matrices according to \( \tau_x(t_m) \mapsto \tau_x^{(k_m)} \) with \( \tau_x^{(k_m)} := (1 - \tau_{k_m} \tau_{k_m-1}) / 2, \tau_y^{(k_m)} := -i(\tau_{k_m} - \tau_{k_m-1}) / 2 \).
and \( \tau_{\xi}^{(k_m)} := \tau_{k_m} \). Since the matrix elements \( \langle \tau' | \tau_{x,y} | \tau \rangle \) are non-zero only for \( \tau \neq \tau' \), the Pauli matrices should be replaced by field expressions that include neighbouring spins. In other words, only if a flip-flop occurs at time \( t_m \), the fields \( \tau_{x,y}^{(k_m)} \) are non-zero. On the forward branch, a flip-flop with \( \tau_k = -\tau_{k-1} \) is associated with time step \( k \). Then, \( S_O[\eta] := \eta O \) and

\[
\langle O \rangle(t_m) = -i \partial_{\eta} \ln \mathcal{Z}[\eta]_{|\eta=0}.
\]

A generalization to observables with two or more time parameters, e.g. correlation functions, is possible via higher-order derivatives of the generating function. We calculate the charge current \( I(t_m) \) via the source term

\[
S_I = -\frac{i e \eta}{2} \sum_{k \sigma p} p \left( \gamma(\mathbf{k}_m) \gamma(\mathbf{k}_m) - \gamma^\dagger(\mathbf{k}_m) \gamma^\dagger(\mathbf{k}_m) \right),
\]

and the expectation value of the impurity \( \langle \tau_\xi(t_m) \rangle \) from \( S_{\tau_\xi} = \eta \tau_{k_m} \).

### 3.5. Tracing out electron degrees of freedom

Next, we perform traces over the lead degrees of freedom and perform the path integral over all Grassmann fields \( \bar{\xi}_{k \sigma p}, \xi_{k \sigma p} \). Contour time integrations are transferred to their respective real-time counterparts. This results in two integrals over real time for the (+) and (−) branches and generates the \( 2 \times 2 \)-matrix structure for the Keldysh Green’s function. The resulting generating function is written as a path integral with the effective electronic dot action \( S_{\text{el}} = S_{\text{el}}^{\text{dot}} + S_{\text{env}} \) with the (effective) environmental action

\[
S_{\text{env}} = \sum_{\sigma p} \int_0^\infty dt \int_{-\infty}^\infty dt' \bar{\sigma}(t) \gamma(p, t-t') \left\{ 1 + \frac{i e \eta p}{2} \left[ \delta_{m}(t') - \delta_{m}(t) \right] \right\} \sigma(t').
\]

Here, we have introduced the time non-local Keldysh matrix

\[
\gamma(p, t-t') = \frac{\Gamma}{2 \beta \sinh[\pi (t-t')/\beta]} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}.
\]

This equation holds only for \( t - t' \neq 0 \). The singularity for \( t = t' \) will be addressed below. The exponential decay of \( \gamma \) for \( |t - t'| \to \infty \) is the cornerstone of the ISPI method, which allows us to truncate certain long-time correlations (see below). Equation (26) can also be given in terms of an environmental Green’s function

\[
S_{\text{env}} = \sum_{\sigma} \int_{-\infty}^\infty dt \int_{-\infty}^\infty dt' \bar{\sigma}(t) \left( G_{\text{env}} \right)_{t,t'}^{-1} \sigma(t') \quad \text{with} \quad \left( G_{\text{env}} \right)_{t,t'}^{-1} = \left( G_{\text{env}} \right)_{t,t'}^{-1} + \eta \left( G_{\text{env}}^{\dagger} \right)_{t,t'}^{-1},
\]

and

\[
\left( G_{\text{env}}^{\dagger} \right)_{t,t'}^{-1} = \frac{e \Gamma \sin[\pi (t - t')/\beta]}{2 \beta \sinh[\pi (t-t')/\beta]} \begin{pmatrix} \delta_{m}(t) - \delta_{m}(t') & -\delta_{m}(t) \\ \delta_{m}(t') & \delta_{m}(t) \end{pmatrix}.
\]

This expression is well defined for all values of \( t \) and \( t' \). An explicit expression for \( \left( G_{\text{env}}^{\dagger} \right)_{t,t'} \) is given below in equation (31).

Before we proceed, we address the \( (t - t')^{-1} \) singularity of \( \left( G_{\text{env}}^{\dagger} \right)^{-1} \). In contrast to its inverse, the Green’s function \( G_{\text{env}}^{\dagger} \) itself is finite at \( t = t' \). Still, matrix elements decay...
Figure 3. \((G_{0,\sigma})^{++}\) versus time difference \(t - t'\) for two combinations of \(\omega_{\sigma}^U\) and e\(V\) for \(\beta \Gamma = 1/5\). Except for \(t = t'\), the function is smooth for all real times. The imaginary part is discontinuous at \(t = t'\), which turns out to be harmless for our numerical scheme. A possible way to cure it is to introduce a high-frequency cutoff \(\exp\{-|\omega|/\omega_c\}\) in equation (32); see dotted lines in the inset for the same vertical scale for \(\omega_c = 100 \Gamma\).

exponentially with growing time differences. The calculation of observables does not suffer from this singularity either, since it cancels in the fraction

\[
\frac{Z[\eta] - Z[0]}{\eta Z[0]} \approx i \langle O \rangle(t_m),
\]

which is used to numerically evaluate \(\langle O \rangle\). Since the divergence depends neither on the paths of the impurity- and the HS-spins nor on \(\eta\), we may rescale \(Z[\eta]\) by the singular factor without affecting \(\langle O \rangle(t_m)\). For clarity, we keep the same notation for the rescaled generating function. The singularity originates from \((G_{\text{env}}^0)^{-1}\). We collect all non-interacting contributions, which do not depend on \(\eta\) into the sum \(\sum_{\sigma} (G_{0,\sigma}^+)^{-1} = \sum_{\sigma} (G_{\text{dot},\sigma}^0)^{-1} + (G_{\text{env},\sigma}^0)^{-1}\) with

\[
(G_{\text{env},\sigma})^{-1} = \sum_p \gamma(p, t - t'),
\]

\[
(G_{\text{dot},\sigma})^{-1} = \delta(t - t') \begin{pmatrix} i \partial_t - \omega_{\sigma}^U & 0 \\ 0 & -i \partial_t + \omega_{\sigma}^U \end{pmatrix},
\]

with \(\omega_{\sigma}^U := (\epsilon_{\sigma} + U/2)\). The Fourier transform of \((G_{0,\sigma})^{-1}\) is obtained as

\[
(G_{0,\sigma})^{-1} = 2\pi \delta(\omega - \omega') \begin{pmatrix} \omega - \omega_{\sigma}^U + i\Gamma[F(\omega) - 1] & -i\Gamma F(\omega) \\ i\Gamma[2 - F(\omega)] & -\omega + \omega_{\sigma}^U + i\Gamma[F(\omega) - 1] \end{pmatrix},
\]

where we have introduced \(F(\omega) = f_L(\omega) + f_R(\omega)\) as the sum of the two lead Fermi distributions. This 2 × 2-matrix is inverted algebraically and transformed back into time space by complex contour integration [53] or numerical integration. The function \((G_{0,\sigma}^{\text{el}})^{++}(t - t')\) is shown in figure 3. Whenever this divergence arises, we will remove it, see for instance equation (42), by the multiplication of \(-i \det G_{0,\sigma}^{\text{el}}\), or equivalently, we replace \(i(G_{\sigma}^{\text{eff}})^{-1}\) with

\[
D_\sigma[\eta] := G_{0,\sigma}^{\text{el}}(G_{\sigma}^{\text{eff}})^{-1} = 1 + \sum_{\sigma} \sigma \Sigma_\sigma^0 + \eta \Sigma_\sigma^U,
\]
with the self-energy
\[
(\Sigma^0_{\sigma})_{kl} = \delta_{kl} \left( -J \tau_k - i \zeta_k \lambda(\delta_l) \begin{array}{cc} 0 \\ J \tau_l - i \zeta_l \lambda^*(\delta_l) \end{array} \right) \delta_l.
\] (34)

The particular form of \( \Sigma^0_{\sigma} \) depends on the observable \( O \). When \( O = I \), we identify it with \((G^\text{env})^{-1}\) of equation (29) and otherwise with \((G^O)^{-1}\).

The discrete form of the matrix \( D_\sigma \) follows via the relation
\[
(G^\text{el}_{0,\sigma})_{kl} = \frac{1}{\delta_l^2} \int_{t_{l-1/2}}^{t_{l+1/2}} dt' \int_{t_{l-1/2}}^{t_{l+1/2}} dt (G^\text{el}_{0,\sigma})_{t,t'} \approx (G^\text{el}_{0,\sigma})_{t,0} \quad \text{with} \quad t_k = t_1 + (k - 1) \delta_l.
\] (35)

To remove the singularity at \( k = l \), we choose the regularization
\[
(G^\text{el}_{0,\sigma})_{ll} = [(G^\text{el}_{0,\sigma})_{(l-l')\rightarrow 0} + (G^\text{el}_{0,\sigma})_{(l-l')\rightarrow 0}]/2.
\] (36)

Alternatively, introduction of a high-frequency cutoff \( \exp[-|\omega|/\omega_c] \) in the Green’s function \([48]\) yields a consistent result; see the inset of figure 3(b). For \( \omega_c U = 3 \Gamma \) and \( eV = 4 \Gamma \), equation (36) yields \((G^\text{el}_{0,\sigma})_{ll} \approx 0.3384i \) and with the cutoff method with \( \omega_c = 100 \Gamma \), we obtain 0.3245i (the difference of \( \sim 4\% \) decreases for larger \( \omega_c \)). However, using equation (36) has two advantages: firstly, it does not modify off-diagonal Green’s matrix elements and, secondly, we do not need an additional parameter \( \omega_c \).

We emphasize that both methods of regularization obey the necessary causality relation, \((G^\text{el}_{0,\sigma})^{++}_{ll} + (G^\text{el}_{0,\sigma})^{--}_{ll} - (G^\text{el}_{0,\sigma})^{+\,}_{ll} - (G^\text{el}_{0,\sigma})^{-\,}_{ll} = 0\). This follows from the causality structure of the Green’s matrix and the self-energies \( \Sigma \) \([52]\). Here, the diagonal elements of both \( G^\text{el}_{0,\sigma} \) and the \( \Sigma \) matrices have to be understood as the average, i.e. the integral of the time non-local matrix elements in an interval \( \delta_l \) around the point \( t - t' = 0 \). The discrete version of \( \Sigma^0_{\sigma} \) follows accordingly. For the current, for example, the use of equation (29) yields
\[
(\Sigma^0_{\sigma})_{kl} = \frac{e \Gamma \delta_l \sin[eV(k-1)\delta_l/2]}{2 \beta} \sin[\pi(k-1)\delta_l/\beta] \left( \begin{array}{cc} \delta_{k,m} - \delta_{l,m} & -\delta_{k,m} \\ \delta_{l,m} & 0 \end{array} \right).
\] (37)

We note that source terms for observables \( O \) containing dot fields may also be added to the action. The effective full inverse Green’s function \((G^\text{eff})^{-1}\) is given by either \((G^\text{dot})^{-1} + (G^\text{env})^{-1}\) for the current or \((G^\text{dot})^{-1} + \eta(G^O)^{-1} + (G^0)^{-1}\) for other observables. Plugging in those pieces, the remaining path integral is recast as a discrete sum, i.e.
\[
\mathcal{Z}[\eta] = \sum_{\{r, \xi\}} \int \mathcal{D}[\tilde{\sigma}_r, \sigma_r] \mathcal{P}[\{\tau\}] \exp \left\{ \int_0^1 \sum_{\sigma} \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt' \tilde{\sigma}_r(t) (G^\text{eff}_\sigma)^{-1}_{t,t'} \sigma_r(t') \right\}
\]
\[
= \sum_{\{r, \xi\}} \mathcal{P}[\{\tau\}] \prod_{\sigma} \det \{ (\sigma G^\text{eff}_\sigma)^{-1}_{\{\tau, \xi\}, \eta} \}. \qquad \text{(38)}
\]

Hence, we have obtained the Keldysh partition function as a sum over expectation values of the polynomial \( P \) of Grassmann numbers in a system with Green’s function \( G^\text{eff}_\sigma \). The next step is to derive an expression for \( \langle P[\{\tau\}] \rangle \) that refers to \( G^\text{eff}_\sigma \) only by applying Wick’s theorem \([52]\). With equation (22), we have
\[
\langle P[\{\tau\}] \rangle = \left( \prod_{j \in T^+_{\text{up}}} \tilde{\sigma}_j^{l+1} \sigma_j^l \prod_{k \in T^+_{\text{up}}} \tilde{\sigma}_k^{l-1} \right) \left( \prod_{j \in T^-_{\text{up}}} \tilde{\sigma}_j^l \sigma_j^{l-1} \prod_{k \in T^-_{\text{up}}} \tilde{\sigma}_k^{l+1} \right).
\] (39)

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For an odd number $m$ of flip-flops the expectation value vanishes, since each process contributes a creator and an annihilator for electrons with opposite spins. Odd $m$ implies an odd number of alternating products of creators and annihilators; the number of $\delta$ is different from the number of $\sigma$. When applied to any state $|\psi\rangle$ in the trace, this changes the particle count by 1 and the projection with $\langle\psi|$ vanishes. Therefore we have to consider paths with even $m$ only.

As an example, we evaluate equation (39) for the impurity path shown in figure 4 before we turn to the general formalism and arbitrary paths $\{\tau\}$. The exemplary path features $m = 4$ flip-flops and we find that

$$
\langle P \rangle = \begin{pmatrix}
\delta_{15} \delta_{7} \delta_{14} \delta_{9} \\
\delta_{8} \delta_{7} \delta_{6} \delta_{3}
\end{pmatrix} = \begin{pmatrix}
\delta_{15} \delta_{7} \delta_{14} \delta_{9} \\
\delta_{8} \delta_{7} \delta_{6} \delta_{3}
\end{pmatrix} = \begin{pmatrix}
\delta_{14} \delta_{9} \delta_{6} \delta_{3} \\
\delta_{8} \delta_{7} \delta_{6} \delta_{3}
\end{pmatrix} = \begin{pmatrix}
\delta_{14} \delta_{9} \delta_{6} \delta_{3} \\
\delta_{8} \delta_{7} \delta_{6} \delta_{3}
\end{pmatrix}.
$$

(40)

$S_{\text{eff}}$ and the expectation value of the mixed operator product factorizes with respect to the spin degree of freedom. Even in the presence of spin-mixing terms this factorization remains valid. Applying Wick’s theorem to equation (40) yields

$$
\langle P^\sigma \rangle = -\det \Xi_{\sigma},
$$

and

$$
\langle P^\dagger \rangle = -\det \begin{pmatrix}
(G_{\text{eff}}^\dagger)_{2,7} & (G_{\text{eff}}^\dagger)_{2,15} \\
(G_{\text{eff}}^\dagger)_{8,7} & (G_{\text{eff}}^\dagger)_{8,15}
\end{pmatrix}
$$

and

$$
\langle P^\dagger \rangle = -\det \begin{pmatrix}
(G_{\text{eff}}^\dagger)_{6,3} & (G_{\text{eff}}^\dagger)_{6,9} \\
(G_{\text{eff}}^\dagger)_{14,3} & (G_{\text{eff}}^\dagger)_{14,9}
\end{pmatrix}.
$$

(41)

The general procedure for constructing $\Xi_{\sigma}$ for an arbitrary path $\{\tau\}$ having an even number $m$ of flip-flops is as follows.

1. Construct the flip-flop polynomial $P^\sigma[\{\tau\}]$.
2. Assign indices $q_1 < \ldots < q_m/2$ of annihilator fields $\delta_{\sigma}^q, \ldots, \delta_{\sigma}^{q_m/2}$ that appear in $P^\sigma[\{\tau\}]$ to the rows of $\Xi_{\sigma}$.
3. Assign indices $r_1 < \ldots < r_m/2$ of creator fields $\overline{\delta}_{\sigma}^r, \ldots, \overline{\delta}_{\sigma}^{r_m/2}$ that appear in $P^\sigma[\{\tau\}]$ to the columns of $\Xi_{\sigma}$.

Using the matrix element $(\Xi_{\sigma})_{k,l} = -i[\delta_{\sigma}^k \overline{\delta}_{\sigma}^l] = (G_{\sigma}^{\text{eff}})_{k,l}$, the final expression for the generating function follows as

$$
\mathcal{Z}[\eta] = \lim_{\delta,\zeta \to 0} \sum_{\{\tau,\zeta\}} (-1)^{\delta/2} \left( \frac{J}{\delta} \right)^m \exp[i S_{\text{imp}}] \prod_{\sigma} \det (G_{\sigma}^{\text{eff}})^{-1} \det \Xi_{\sigma},
$$

(42)
where the summation over impurity paths is restricted to tuples \{τ\} with \(τ_1 = τ_2 = ... = τ_i\), i.e. correct boundary conditions along the Keldysh contour are imposed. The limit \(δt \to 0\) appears explicitly here, since there is no continuous measure used for the discrete spin paths, neither for the HS- nor for the impurity spins.

### 3.6. Iterative summation of the path integral

The exact generating function in equation (42) is intractable due to the exponentially growing size of the matrices for long propagation times and an adept numerical treatment is necessary in order to proceed. The off-diagonal elements of \((G_{\text{eff}})^{-1}\), given in terms of \(γ(p, t - t')\) in equation (27), decay exponentially with increasing distance \(t - t'\) from the diagonal, at finite \(T\) and/or \(V\) [48]. This fact is illustrated in figure 5. In addition, the bias voltage induces oscillations. The correlation or memory time \(τ_c =: (K - 1)δ_t\) determines the range of lead-induced correlations; these are exponentially suppressed for time differences larger than \(τ_c\). This allows us to restrict the path summation to a finite memory time window and thus the number of paths that contribute to \(Z[η]\), originating from the magnetic impurity as well as from the Coulomb interaction, is drastically reduced. We use \(K\) as a memory time parameter in the ISPI scheme in an extrapolation procedure for \(τ_c \to ∞\). The latter eventually gives converged results independent of \(τ_c\).

To proceed, we rotate the basis unitarily, thereby rearranging the matrix elements of \((G_{\text{eff}})^{-1}\) such that increasing distances with respect to the diagonal correspond to increasing time differences. For a given \(K\), we obtain the block band-matrix

\[
(G_{\text{eff}})^{-1} \approx \begin{pmatrix}
A_{1,1}^σ & A_{1,2}^σ \\
A_{2,1}^σ & A_{2,2}^σ \\
& \ddots \\
& & & A_{N_c-1,N_c}^σ \\
& & & & A_{N_c,N_c}^σ
\end{pmatrix},
\]

(43)
Figure 6. An example of an impurity path with 12 flip-flops (top) and the associated $\Xi_\downarrow$-matrix (bottom) for $\tau_c = 2\delta t$ ($K = 3$). The discrete path (arrows on the vertices) has a length of $8\delta t$ ($N = 9$) and is divided into $N_C = 3$ segments of length $K$ (separated by dotted lines). Depending on the flip distribution, the diagonal blocks $B_{i,i}$ are, in general, not quadratic (boxes with dashed frames) and their determinants do not exist. To render them quadratic, we reassign the fields of the flips closest to the segment borders (blue arrows). The hatched elements are exponentially small and neglected.

where $N_C = N/K$ and the blocks $[A]_{k,l}^\sigma$ are $K$-dimensional matrices, whose entries are given by those of $(G^{\text{eff}})^{-1}$ taken from the rows and columns in the range of $\{(j-1)K+1, \ldots, jK\}$ with $j = k, l$. Since we neglect exponentially small components, the $[A]$-blocks in the upper (lower) secondary diagonal have an upper (lower) triangular structure. The $\Xi_\sigma$ matrices in equation (42) naturally inherit the same block structure with the modification that the corresponding blocks $[B]_{k,l}^\sigma$ are in general not quadratic. Their dimensions are determined by the number of flip-flops within the respective time interval.

To illustrate the scheme, a particular impurity path is shown in figure 6, which is of length $8\delta t$, consisting of $2N = 18$ vertices having 12 flip-flops. The calculation of the determinant in our scheme needs quadratic block matrices on the diagonal, which we obtain again by a unitary transformation. The off-diagonal blocks are reshaped accordingly. In figure 6, this procedure is illustrated for $\Xi_\downarrow$ with $\tau_c = 2\delta t$ ($K = 3$), obtained after the rearrangement. The hatched matrix elements are disregarded. The path with $N = 9$ is divided into $N_C = 3$ segments with $K$ vertices on each branch. In analogy to the blocks $[A]_{k,l}^\downarrow$, the diagonal blocks $[B]_{i,i}^\downarrow$ contain all matrix elements $G^\downarrow_{iqr}$ with $(i-1)K < q, r \leq iK$ (dashed boxes). Since the number of flip-flops is odd, all the matrices $[B]_{i,i}^\downarrow$ are not quadratic. Instead, this particular case yields $2 \times 1$-matrices.
Neglect in the resulting element of second row, second column products like $\sigma_1 \sigma_2$, 
and perform a Gaussian elimination of the block in the second row, first column.

Expand the determinant after the first column, thus reducing the problem by one in block dimensions.

\[
\begin{align*}
\left[\begin{array}{c}
B_{1,1}^\dagger \text{ and } B_{3,3}^\dagger,
\end{array}\right] \text{ and a } 2 \times 4\text{-matrix } B_{2,2}^\dagger \text{. The quadratic blocks required for the iteration scheme are obtained by reassigning the earliest and latest creator fields } \bar{s}_{i,4}^\dagger \text{ and } \bar{s}_{i,6}^\dagger \text{ from the second segment to the first and third, respectively (blue arrows). Such a reordering is always possible and renders all diagonal blocks of } \Xi_\sigma \text{ quadratic.}
\end{align*}
\]

We define the recursive notation $X = A, B$ to compactify the computation of the determinant for the blocked Keldysh partition function as

\[
X = \begin{bmatrix} X_D \end{bmatrix} \text{ with } X_D = \begin{bmatrix} X_{i,i} & X_{i+1,i} \\ X_{i,i+1} & X_{D-1} \end{bmatrix} \text{ and } X_1 = \begin{bmatrix} X \end{bmatrix}_{D,D}.
\]

The double line denotes matrices which themselves consist of blocks, with the subscript $D$ giving their dimensions in blocks. The determinant of $X$ is calculated iteratively \cite{48} in $D - 1$ steps each of which consists of the following manipulations.

(i) Perform a Gaussian elimination of the block in the second row, first column.

(ii) Neglect in the resulting element of second row, second column products like $\begin{bmatrix} X \end{bmatrix}_{k-1,k}, \begin{bmatrix} X \end{bmatrix}_{k,k+1}$, which connect segments beyond the nearest neighbour.

(iii) Expand the determinant after the first column, thus reducing the problem by one in block dimensions.

While steps (i) and (iii) are exact algebraic operations, step (ii) is the second building block of the ISPI method and is necessary for the scheme to remain consistent with neglecting the correlations beyond $\tau_c$. A step $k \rightarrow k + 1$ is performed solely based on the determinant after step $k$ and the spin orientations in segments $k$ and $k + 1$. Here we stress that within the time window $\tau_c$, the ISPI scheme takes into account important non-Markovian effects in a natural way. In this work, these correlations are lead induced. Within a typical Markovian approximation, the real-time dependence of the Green’s function in figure 6 is replaced by $\delta(t - t')$. To include terms in the iteration that connect segments beyond nearest neighbouring $K$-blocks would require information about impurity spins in ‘earlier’ segments $< k$, which are beyond $\tau_c$. After step (iii), we arrive at

\[
\det X = \det \begin{bmatrix} X \end{bmatrix}_{1,1} \begin{bmatrix} X_{2,2}' & X_{2,3} \\ X_{3,2} & X_{D-2} \end{bmatrix},
\]

where $\begin{bmatrix} X \end{bmatrix}_{2,2}' = \begin{bmatrix} X_{2,2} - X_{2,1}^{-1} X_{1,2} \end{bmatrix}$. Subsequent iteration gives the final relation

\[
\det X = \det \begin{bmatrix} X \end{bmatrix}_{1,1} \prod_{i=2}^{D} \det \begin{bmatrix} X_{i,i} - X_{i,i-1} X_{i-1,i}^{-1} X_{i-1,i-1} \end{bmatrix}.
\]

Next, we construct the $\Xi_\sigma$-matrices for finite correlation times starting from equation (42) by filling the entries with the elements of $G^{\text{eff}}_\sigma(\tau, \zeta)$. The latter depends on the entire spin path $\{\tau, \zeta\}$ between $t_i$ and $t_f$. In principle, inversion of the full inverse Green’s function is possible but is out of the reach for practical applications since the numerical effort grows exponentially with propagation time. To remain consistent with the finite correlation time approach, we have to find approximations of $\begin{bmatrix} B \end{bmatrix}_{i,1(\pm 1)}$ that depend on spins of the nearest neighbour segments $i(\pm 1)$.
This allows us to construct the $B$-blocks for all $1 \leq k = 6$ spins. For example, the path segment $\{\tau\}_l = (\tau_j^-, \tau_j^+, \tau_j^-, \tau_j^+, \tau_j^+)$ ($\downarrow$, $\uparrow$, $\downarrow$, $\uparrow$, $\downarrow$). Accordingly, we find that $\{\zeta\}_3 = (\zeta_9^-, \zeta_9^+, \zeta_8^-, \zeta_7^+, \zeta_7^+)$ ($\uparrow$, $\downarrow$, $\uparrow$, $\downarrow$, $\downarrow$). The compact notation combines the impurity- and HS spins as $\{\tau, \zeta\}$, e.g. $\{\tau, \zeta\}_2 = (\uparrow, \downarrow, \downarrow, \uparrow, \downarrow, \downarrow, \uparrow, \downarrow, \downarrow)$. We observe that blocks on the secondary diagonals contribute much less than those on the main diagonal. Hence, we expand $(G^\text{eff})^{-1}$ in powers of the off-diagonal blocks, yielding

$$G_{k,k} \approx A_{k,k}^{-1} \quad \text{and} \quad G_{k,l} \approx -A_{k,k}^{-1}A_{k,l}A_{l,l}^{-1},$$

for all $1 \leq k, l \leq N_C$ and $|k - l| = 1$ (the index $\sigma$ was omitted). The blocks $G_{k,j}$ are defined in analogy to the $A$-blocks and the $B$-blocks are filled as described above.

Next, we use from equation (33) the relation $G^\text{eff} = D^{-1}G^\text{el}_{0,\sigma}$, see section 3.5. The $\Xi$-matrices are also free of any singularity, and from equation (47), we obtain the approximate inverse of $D_{\sigma}$. We multiply the result by the free Green’s matrix in block form. For the step $k - 1 \rightarrow k$, we obtain

$$G_{k-1,k-1} = D_{k-1,k-1}^{-1} \left[ G_{0,k-1,k-1} - D_{k-1,k-1}^{-1} D_{k-1,k}^{-1} G_{0,k,k}^{-1} \right],$$

$$G_{k-1,k} = D_{k-1,k}^{-1} \left[ G_{0,k,k-1} - D_{k-1,k}^{-1} D_{k-1,k}^{-1} G_{0,k,k}^{-1} \right],$$

$$G_{k,k} = D_{k,k}^{-1} \left[ G_{0,k,k} - D_{k,k}^{-1} D_{k-1,k-1}^{-1} G_{0,k-1,k-1}^{-1} \right].$$

This allows us to construct the $B$-blocks without calculating the inverse Green’s function explicitly.

Collecting all parts, we can finally express $Z[\eta]$ iteratively as

$$Z[\eta] = \sum_{\{\tau, \zeta\}} Z_{N_C}, \quad \text{where} \quad Z_j = \sum_{\{\tau, \zeta\}_{j-1}} \Lambda_{j-1,j} Z_{j-1}. \quad (49)$$

The real-time propagator of the ISPI scheme is introduced as

$$\Lambda_{j,j-1} = F_j \prod_{\sigma} \prod_{X=H,D} \det \left[ X_{j,j}^{\sigma} - X_{j,j-1}^{\sigma} X_{j-1,j-1}^{\sigma} X_{j-1,j}^{\sigma} \right],$$

$$\text{New Journal of Physics} 14 (2012) 073049 (http://www.njp.org/)$$
with the chosen initial configuration
\[ Z_1 = F_1 \prod_{\sigma} \prod_{X=B,D} \text{det} X_{1,1}^\sigma. \] (51)

Furthermore, we use the definition \( \{ \tau, \xi \}_j = (\tau_{j;K}, \ldots, \tau_{j(K+1)}, \xi_{j;K}, \ldots, \xi_{j(K+1)}) \) as the tuple of those impurity and HS spins that lie in the \( j \)th path segment of length \( K \), see figure 7. The propagator \( \Lambda_{j,j-1} \) (and matrix blocks) depends on all HS and impurity spins in segments \( j \) and \( j-1 \). The prefactor
\[ F_j = 2^{-2K} (-1)^{\ell_j} \left( \frac{J_1}{2} \right)^{m_j} \exp^{\phi_{\text{imp}}^{(j)}} \] (52)
is related to the number and the position of the flip-flops. \( m_j \) is the number of flip-flops in segment \( j \), out of which \( \ell_j \) lie in the backward branch. The phase is defined as
\[ \phi_{\text{imp}}^{(j)} = \frac{-\Delta_{\text{imp}} \delta_t}{2} \sum_{l=(j-1)K+1}^{jK} (\tau_l^+ - \tau_l^-). \] (53)

3.7. Extrapolation procedure

By construction, the numerical value of \( Z[\eta] \) contains two systematic errors, namely the finite discretization time step \( \delta_t \) and the finite correlation time \( \tau_c = (K-1) \delta_t \). In the limit \( \delta_t \to 0 \) and \( K \to \infty \), however, the iterative procedure yields an exact result. A major benefit of this iterative scheme is that the numerical costs scale linearly with the evolution time \( t_m - t_i \). The systematic errors are eliminated [48] by an extrapolation of the numerical results to vanishing Trotter increment \( \delta_t \) and infinite memory time \( \tau_c \to \infty \).

Due to the Trotter time discretization, all expressions are by construction exact up to order \( \delta_t^2 \) terms. For a fixed \( \tau_c \) and small enough values of \( \delta_t \), we extrapolate the numerical values of some observable to \( \delta_t \to 0 \) and thus completely eliminate the Trotter error. An example is shown in figures 8(a) and (b) for the current and the impurity orientation, respectively. Depending on the observable, Trotter convergence may be achieved on different scales [57]. Note that one source of errors is the numerical derivative in equation (30), which results in tiny imaginary parts of observables, ranging between \( 10^{-5} \) and \( 10^{-3} \). For typical parameters, it is at least one order of magnitude smaller than the numerical error from the linear extrapolation.

For the memory extrapolation \( \tau_c \to \infty \), we do not have a strict mathematical argument at hand, in contrast to the Trotter extrapolation. Whenever results are convergent, however, we find empirically two typical behaviours: either (i) the numerical results for \( \langle O \rangle (\tau_c) \) depend linearly on \( 1/\tau_c \) with small deviations or (ii) their dependence on \( 1/\tau_c \) is reasonably smooth and exhibits a local extremum. The latter case is consistent with the principle of least dependence; see [48, 49, 54, 55] for a verification when compared to analytical results. An example of the linear scaling of the numerical results with \( \tau_c^{-1} \) is illustrated in figures 8(c) and (d). When \( \langle O \rangle (\tau_c^{-1}) \) shows a weak dependence on \( \tau_c^{-1} \) in a certain corner of the parameter space, we still try to apply criterion (ii). Such a behaviour results from a trade-off between accuracy and computational costs. A minimal Trotter error requires minimal \( \delta_t \), while, at the same time, a maximally large correlation time \( \tau_c \) is desirable. Naturally, these requirements are limited by the exponentially increasing numerical costs. This is illustrated in figure 9.
3.8. Restricting the number of flip-flops in the memory window

In order to reduce the exponentially growing number of contributing paths ($\sim 4^K$) without affecting the accuracy, we may exploit that $F_j \sim (J \delta_t/2)^{m_j}$ in equation (52). The number $m_j$ of flip-flops in path segment $j$ is $0 \leq m_j \leq 2(K - 1)$. We observe that the smaller the weight of each segment, the more flip-flops it contains. On the other hand, the number of path segments $\{\tau_j\}$ with $m_j$ flip-flops (given by $4C_{m_j}^{2(K-1)}$ with $C_n^k = n!/[k!(n-k)!]$) grows as long as $0 \leq m_j \leq K - 1$, but decreases again when $K \leq m_j \leq 2(K - 1)$. As a consequence, for any observable there exists a maximal $m_j^{\text{max}}$ such that the contributions from paths with $m_j > m_j^{\text{max}} \leq 2(K - 1)$ could safely be disregarded in the numerical iteration. Of course, $m_j^{\text{max}}$ is chosen depending on the model parameters and the observable under investigation.

Rapidly decreasing weights of the paths may not be (over-)compensated by increasing weights for $0 \leq m_j \leq K - 1$, since each contribution is small and the number of paths decreases again for larger $m_j \geq K$. The behaviour of the impurity weights is illustrated as follows.

**Figure 8.** (a, b) Trotter extrapolation (solid lines) for $\delta_t \to 0$ for $\Gamma_c = 1/\Gamma$, $t_m = 4$, $\beta = 1$ and $\Phi_0 = \Delta = \Delta_{\text{imp}} = 0$; (a) charge current in units of $I_0 = e\Gamma/h$ for $J = 0$, (b) mean impurity spin polarization for $eV = 0.5\Gamma$ and $U = 0.5\Gamma$. The impurity spin was initially in the spin-up state $\tau_i = 1$. Tiny deviations point to negligible unsystematic errors. For all the cases in (a), the standard deviations are below 1%, while in (b), they rise from around 1% for $J = 0.3\Gamma$ to about 10% for $J = 0.9\Gamma$. (c, d) Memory extrapolation $\tau_{c-1} \to 0$ for (c) the current and (d) the impurity orientation. For all combinations of $eV$ and $U$, the standard deviations to the linear fit are below 1%. Within the error margin, the numerical value of the current for $eV = 0.6\Gamma$ and $U = 0$ coincides with the Landauer–Büttiker (LB) value $I_{\text{LB}} \approx 0.594 I_0$. 

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Consider the case when $m_j$ is close to the maximum $2(K - 1)$. Both path classes with $m_j = 0$ and $m_j = 2(K - 1)$ contain the same number of elements (four), while each path contribution in the second class is weighted by $(J\delta_t/2)^{2(K-1)}$. For typical values of $K = 4$, $\delta_t \Gamma = 1/2$, and $J = \Gamma$, the weight is $\sim 2.5 \times 10^{-4}$. This also holds for all $K \leq m_j \leq 2(K - 1)$. Since $m_j^\text{max}$ is unknown a priori, we include it in our code as an additional parameter. Then, we perform a numerical estimate by a spot sample of the parameter space. It turns out that for many cases, it is sufficient already to choose $m_j^\text{max} = 2$. Figure 10 shows an example where both $\langle I \rangle$ and $\langle \tau_z \rangle$ converge quickly for increasing $m_j^\text{max}$. This drastically reduces the CPU running times, e.g. for parameters chosen as in figure 10, from more than one month to typically three to five days.

4. Charge current and impurity dynamics

The ISPI scheme was originally developed for the Coulomb-interacting SLQD. By reproducing established analytical and experimentally confirmed results, the general validity of the approach has been shown in [48, 49]. Here, we focus on novel transport features caused by the magnetic impurity and its interaction with dot electrons. We emphasize that novel dynamical and transport features are mediated by the transverse or flip-flop interaction $\hat{H}_\text{int}^\perp$, given by equation (4). Without the possibility of flip-flops the orientation of the impurity spin and its quantum state could not change and not participate in the dynamics. The remaining longitudinal part of the interaction $\hat{H}_\text{int}^\parallel$ causes a renormalization of rates and energies, which adds to the effect stemming from a magnetic field. Necessarily, flip-flop processes are included in the calculations.

Figure 9. (a) Mean impurity polarization versus $\tau_z^{-1}$ for different measurement times $t_m$ for $\Phi_D = -\Gamma/2$, $\Delta = \Gamma/2$, $J = -\Gamma/2$, $\beta \Gamma = 5$ and $\Delta\text{imp} = U = 0$. Note that the vertical scale is different for each $t_m$ to stress the relative variations. For all plots, the lower bound of the scale is set to $\tau_z^\text{min} = 0.025$, while the upper bound $\tau_z^\text{max}$ is as indicated (the solid lines are a guide to the eyes). The dashed line illustrates the results for $t_m \Gamma = 80$ (green triangles) when scaled as the data for $t_m \Gamma = 4$ (blue crosses). The optimal values of $\langle \tau_z \rangle$ evaluated at the local minimum are shown in panel (b) as a function of $t_m$. A fit to an exponential with a relative standard deviation of $5 \times 10^{-3}$ is shown by the solid line.
from the beginning to investigate the non-trivial impurity dynamics by considering the time dependence of the impurity orientation $\langle \tau_z \rangle$.

In all the results presented below, the impurity is initially polarized and then the coupling to the leads is switched on. We observe that the time decay of the polarization is well described by a single exponential with a constant decay rate. In order to single out the relevant physical processes, we compare our numerical ISPI results to a diagrammatic perturbation theory in the weak to intermediate exchange interaction regime.

We show that for the appropriate parameter regime, the exact numerical results are in agreement with the perturbative result and, by this, we obtain a first intuitive explanation of the impurity dynamics. A next step is the transfer of its plausibility to the ISPI results. However, interaction-induced deviations from the perturbative theory are large enough to clearly illustrate the need for a non-perturbative theoretical description, provided by the ISPI results.

4.1. Real-time decay of the impurity polarization

In figure 11, we present the time evolution of the impurity polarization $\langle \tau_z \rangle$ for different values of the exchange interaction $J$. The remaining model parameters are chosen as $\Phi_D = \Delta = \Delta_{\text{imp}} = U = 0$ and $\beta \Gamma = 1$ as well as $eV = 0.6 \Gamma$. As a function of time, the impurity polarization shows a decaying behaviour, well described by an exponential relaxation for intermediate to long propagation times. A faster decay of the polarization is observed as the impurity interacts stronger with the electron spins. In passing, we note that a rate equation ansatz with a constant rate $\tau_R^{-1}$, where $\tau_R$ is the relaxation time, also results in an exponential decay, i.e. $\langle \tau_z \rangle(t) \propto e^{-(t-t_i)\tau_R^{-1}}$. The parameters are chosen to yield an isotropic (symmetric with respect to (relative) spin orientations) model system. In this case the anti-ferromagnetic interaction favours anti-parallel orientation of electron and impurity spin. Over long propagation times, the coupling to the unpolarized leads then destroys any polarization of the impurity. It is therefore reasonable to assume that the rates for up- and down-flips are equal. In the chosen parameter range, the polarization of the impurity in contact with the leads is described well by a Markovian dynamics, i.e. solely by the time-dependent probabilities $P_r(t)$ of finding the impurity in state $|\tau \rangle$ at time $t$. Apparently, this simple theoretical prediction agrees well with the numerical
Figure 11. The expectation value $\langle \tau_z \rangle$ (log scale) of the impurity orientation as a function of time for four different strengths $J$ of an anti-ferromagnetic electron–impurity interaction. The initial preparation of the system at $t = -\infty$ is spin-up [$\tau_z(0) = \tau_i = 1$] and we set $\Phi_0 = \Delta = \Delta_{\text{imp}} = U = 0$, $\beta = 1$ and $eV = 0.6\Gamma$. The calculated impurity orientation (plot marks) is fitted with good accuracy (errors indicated by shaded areas) to exponentially decaying functions (solid lines).

results, see figure 11. While the impurity interaction energy is comparable to the tunnel coupling and strongly affects the transport behaviour as we show below (see figure 13), the rather high temperature and bias voltage nevertheless reduce the relevance of coherent dynamics due to on-dot interactions to a secondary role.

We then turn to the investigation of the inverse relaxation time $\tau_{\text{R}}^{-1}$. In figure 12(a), we present the results for varying $J$ and $U = 0$ and three different bias voltages. These show a nearly quadratic behaviour growing from zero (no relaxation) in the sense that for a fit of the results for $0 \leq J \leq \Gamma/2$ to a polynomial function $a J^b$ the exponent $b$ lies between $\sim 1.8$ and $\sim 1.9$. An exact quadratic dependence of $\tau_{\text{R}}^{-1}$ on $J$ is obtained only in the cases where the dynamics is strongly dominated by sequential (incoherent) flip-flop processes. This is realized only when $J \ll \Gamma$. The corresponding rates may be obtained based on the real-time diagrammatic technique developed by Schoeller and Schön [24], yielding

$$
\tau_{\text{R}}^{-1} \approx \frac{J^2 \Gamma^2}{16\pi} \sum_{\alpha,\alpha'} \int_{-\infty}^{\infty} d\omega \frac{[f_L^+(\omega) + f_R^+(\omega)][f_L^-(\omega) + f_R^-(\omega)]}{[(\omega - \omega_\uparrow + \alpha J)^2 + \Gamma^2][(\omega - \omega_\downarrow + \alpha' J)^2 + \Gamma^2]}.
$$

Details of the derivation can be found in [53]. It reveals the physical structure and allows for the intuitive interpretation of the processes contributing to sequential flip-flops. In the numerator of the integrand, we have the sum of all four possible ways to multiply one of the lead’s occupations ($f^+$) with another or the same lead’s probability of finding an empty state ($f^-$) at some energy. Each of these four combinations is then multiplied by the Lorentzian spectral density for the two different spin states each shifted by $\pm J$ (longitudinal interaction energy). This suggests the following interpretation: a sequential flip-flop process consists of three elementary components: the actual flip-flop and two tunnelling processes of single electrons with opposite spin (not necessarily in that order). Since they evolve coherently, these components form an effective spin-flip process $|\chi, \tau\rangle \rightarrow |\chi, -\tau\rangle$, where $\chi \in \{0, \sigma, d\}$ and the underlying flip-flop nature is
Figure 12. (a) Impurity relaxation rate $\tau_R^{-1}$ versus interaction strength $J$ for three different values of $V$. The solid lines are fits to polynomial functions $aJ^b$ and all the resulting values of $b$ are close to $2$ ($1.8 \leq b \leq 1.9$). The polarization decays faster with increasing $J$. (b) Comparison of the numerically exact (ISPI, crosses) and the sequential relaxation rate (equation (54), blue solid line) versus bias voltage for $J = \Gamma/2$ and $\beta\Gamma = 1$ (here and in what follows, the parameters not given explicitly are set to zero). Quantitatively, the sequential and ISPI relaxation times differ noticeably in the crossover regime.

masked by the tunnelling electrons. For a particular choice of $\alpha$ and $\alpha'$ in equation (54), we assign certain effective flip processes.

Figure 12(b) shows how the ISPI result (blue crosses) compares to the sequential relaxation time (blue solid line). Although the latter is of the correct order of magnitude, it is systematically larger than the exact value by $\gtrsim 10\%$. Since $J$ is not a small parameter of the system, we can presume that the deviations are mostly coming from coherent higher-order flip-flop processes, which are neglected in equation (54). Another source of those deviations may be that free Green’s functions are used for the derivation of the rates in equation (54). In their qualitative features, however, both the results agree. From their finite value at zero bias voltage they grow monotonically, whereas for small voltages the relaxation shows a nearly quadratic functional form (power law), for large bias voltages $eV \gtrsim 1.25\Gamma$ it exhibits a linear behaviour.

4.2. Impact of the impurity interaction on the current

As opposed to the slow impurity dynamics, measured in terms of $\Gamma^{-1}$, the current is relaxing fast into the stationary state. This behaviour is caused by the strong coupling between the leads and the dot. For the parameters considered here, the upper limit for reaching stationarity is about $t_{\text{ST}} \lesssim \Gamma^{-1}$. Therefore, we consider the stationary current. Figure 13 depicts the current as a function of $J$ with $V = 0.6\Gamma$ and $\beta\Gamma = 1$ both for vanishing Coulomb interaction and for $U = \Gamma/2$. The current decreases with stronger impurity interaction. To distinguish the influence of the longitudinal (single-particle) and the transversal (spin scattering) part of the interaction, we compare the ISPI results with the LB current $\langle I \rangle_{\text{LB}}$ (see [56]), which can be written here as

$$\langle I \rangle_{\text{LB}} = \frac{e\Gamma^2}{2\hbar} \sum_{\sigma,\alpha=\pm} \int_{-\infty}^{\infty} d\omega \frac{f^+_{\sigma}(\omega) - f^+_{\sigma}(\omega)}{(\omega - \omega_{\sigma} + \alpha J)^2 + \Gamma^2}.$$ (55)
Figure 13. Stationary charge current $\langle I \rangle$ in units of $I_0 = e\Gamma/h$ at $eV = 0.6\Gamma$ against $J$ for two values of $U$. For increasing interaction, both the ($U = 0$) LB theory and the numerics predict a decrease of the current in the range $0 \leq J \leq \Gamma$. The similar characteristics of the LB curve and the ISPI data points suggest that the current is mainly affected by the longitudinal part of the electron–impurity interaction. This is probably due to the relatively high temperature and, consequently, a short coherence time, which strongly limits the influence of coherent dynamics. While the LB theory and ISPI agree for $J = \Gamma/8$, growing differences for increasing $J$ show the effect of flip-flop processes. The current for small finite Coulomb interaction (no error bars given), although consistently smaller than the LB values and also decreasing with $J$, drops more slowly than for vanishing $U$.

For $J \neq 0$ this is an approximate expression, as it only includes the effect of the longitudinal impurity interaction, which acts as an effective magnetic field in the sense of a mean field. Similar to the sequential relaxation rates of equation (54), the current formula has a simple physical interpretation. The joint density of dot-electron states is given by a Breit–Wigner function, whose width equals the tunnel coupling strength and whose resonance lies at the single-electron energy $\omega_{\sigma \pm J}$. Hence, the (non-interacting) current is given by the integral over the energy-dependent difference of the left and right lead’s occupation multiplied by the density of the available dot states at that same energy. The difference in occupation of the lead electronic states is largest around the Fermi level, where it has the biggest overlap with the density of states for $J = 0$. With increasing $J$, the density resonances ‘move away’ from the Fermi level, where $f_{L}(\omega) - f_{R}(\omega)$ decreases and the current drops. This effect is explained in terms of the single-particle energy shift due to the longitudinal component of the impurity interaction only.

Figure 13 shows that the flip-flop term $\hat{H}_{\text{int}}$ has a much smaller influence on the charge current at this rather large temperature (incoherent regime) than the longitudinal part of the interaction. Despite the qualitatively similar behaviour of the LB current and the exact data, the flip-flop scattering causes an additional significant current drop that grows for growing $J$. A finite Coulomb interaction of $U = \Gamma/2$ increases the resistivity of the dot and the ISPI points are consistently lower than the LB values. The decreasing effect of the longitudinal part of the impurity, however, is partially compensated for by a broadening of the joint density of states due to Coulomb fluctuations.
4.3. Finite impurity interaction and Coulomb repulsion

In the deep quantum regime, where no small parameter exists, ISPI is certainly applicable and able to describe physical properties not predictable by perturbative methods. In this section, we study how the relaxation rate and the current behave as functions of bias voltage, Coulomb interaction and temperature, respectively.

Figure 14 presents the results in the voltage range $0 \leq eV \leq 3\Gamma$, with $J = \Gamma$, $U = \Gamma/2$ and for temperatures $\beta\Gamma = 1, 2$ and 5. The ISPI data of $\tau^{-1}$ are indicated by symbols, while the solid lines mark the corresponding perturbative rates. The latter exhibit the same features (power-law growth, followed by a (quasi-)linear behaviour, which finally saturates) as in figure 12(b), which are more pronounced for lower temperatures. As expected, the ISPI data points deviate considerably from this lowest-order approximation. Quantum coherent effects are increasingly relevant since all energies are of the same order. Both the degree of quantitative differences and the deviations in the qualitative behaviour increase for lower temperatures.

In figure 15, for each of the two different temperatures, four different current curves are shown—one for each possibility to either have (i) only mean-field dynamics (LB), (ii) the full Coulomb interaction without flip-flop processes (‘no flips’), (iii) flip-flop dynamics without Coulomb fluctuations (‘mean-field $U$’) or (iv) the fully interacting dot (‘full int.’). For $J = \Gamma$ and $V = 2\Gamma$, the Coulomb energy is varied between $0 \leq U \leq \Gamma$. The situation ‘mean-field $U$’ is implemented by setting $\Phi_D = U/2$ and the HS parameter $\lambda = 0$ to illustrate the effect of the ‘classical’ or mean-field part of the Coulomb interaction. This is tantamount to setting the third term in equation (11) to zero, thereby neglecting the Coulomb interaction-induced fluctuations, which results in a shift of each single-electron energy by $U/2$. Since this shift tends to move the transport channels away from the Fermi level, i.e. the region with the highest density of state in the leads, the current drops. By its nature, this decrease is equivalent to that observed for the LB current.
Figure 15. Comparison of (i) the LB current (dashed lines), (ii) the Coulomb interacting current without flip-flop scattering (‘no flips’), (iii) the current without Coulomb scattering but full impurity interaction (‘mean-field $U$’, see text) and (iv) the fully interacting current (‘full int.’) regarding their dependence on the Coulomb interaction $U$ for (a) $\beta \Gamma = 1$ and (b) $\beta \Gamma = 5$. The other (non-zero) parameters are $J = \Gamma$ and $V = 2\Gamma$. The red-shaded areas indicate the error margin for the case ‘mean-field $U$’.

Only for the ‘single-interaction’ currents (‘no flips’), we show the error bars. The reason why no margin of confidence is given for the fully interacting case, regards the comparability of the error data. Calculating the ‘full int.’ current is a very time-consuming task and thus the extrapolation involves considerably fewer data points. Nevertheless, this does not render these values unreliable. We still see a compelling linear behaviour of the $1/\tau_c$ extrapolation with errors of the order of 1% based on the sample standard deviation. Note that with about 10%, the relative error of the current values is rather small; the small variations of the ‘full int.’ data are solely due to the extrapolation errors and have no physical meaning.

For both temperatures, both the LB current and the current without Coulomb scattering show only a weak dependence on $U$ owing to the single-particle energy shift. The current with full Coulomb interaction but a fixed impurity shows a local maximum for $U \sim \Gamma/2$, which is more pronounced for $\beta \Gamma = 5$. In this case, the fixed impurity acts as an effective static magnetic field. The ISPI values for the fully interacting dot vary strongly over the considered $U$ interval, but are scattered around the ‘no flips’ and ‘no $U$’ curves.

As long as the Coulomb interaction is small, all current values in both the figures lie close. The difference between the respective current values is given by the inclusion or exclusion of flip-flop processes alone. Hence, the rather good agreement of the $U = 0$ values suggests that even at this temperature, flip-flop processes alone can affect the current only weakly. Nevertheless, for lower temperatures, the flip-flop processes start influencing the current more strongly, which results in an increased resistivity. The case of the ‘no flip’ current (fixed impurity) is equivalent to a Coulomb-interacting SLQD in a magnetic field. Both curves in panels (a) and (b) show a very similar dependence on $U$, featuring a local maximum at around $U = \Gamma/2$. For the lower temperature, the relative height of the broad current peak is twice as large as for $\beta \Gamma = 1$. This effect is also caused by the broadening of the dot’s joint density of states due to the Coulomb fluctuations.
5. Conclusions

We have studied the real-time non-equilibrium dynamics of a single-orbital magnetic QD including Coulomb interactions. In order to obtain stationary non-equilibrium states at asymptotic times, the ISPI scheme is employed and extended to the cases when an additional magnetic degree of freedom is present. Besides the appearance of an HS field, which decouples the Anderson repulsion term in the Hamiltonian, we have to include the impurity interaction at the same level. This non-trivial task requires an additional summation over paths of the impurity spin degree of freedom. The resulting action in the path integral formalism involves the Green’s function of the QD as well as its inverse. Inversion of the Green’s matrix enlarges the numerical effort tremendously. From the technical point of view, the appearing matrices dealing with the impurity dynamics may violate the necessary block diagonal structure. However, a unitary transformation helps us to build up proper ingredients for the algorithm. Then, impurity-induced correlations also become tractable and do not violate the exponential decay of quantum many-body correlations. We have presented how an efficient truncation scheme provides accurate results for the coupled spin dynamics. Results are given for a quantum spin-1/2 impurity on the dot, whereas the generalization to an impurity with a larger spin is possible. This would be necessary when, e.g., a Mn system is under investigation.

For the same kind of exchange coupling, the implementation of a Mn impurity essentially increases the dimension of the impurity path sum. Instead of summing over all step-like paths of a spin-1/2, it involves the sub-class of step-like paths (steps between orientations differing by one) in the space of the six possible orientations of spin 5/2. Of course, the numerical efforts also increase but are still within the reach of the ISPI method. Further work devoted to this goal will be carried out.

The exponential drop of time correlations due to the leads’ coupling allows for an efficient truncation of the appearing sums—the main building block of the ISPI scheme. Its application yields high-quality numerical data for the impurity relaxation time and the tunnelling current as a function of the bias voltage in the presence of (Coulomb) and electron–impurity interactions. We have performed the necessary checks to compare our findings with established results. In the regime of small impurity interaction, where sequential flip-flops dominate the impurity dynamics, we found good agreement with a classical rate equation. This is a useful tool to gain insight into the dominating processes in the incoherent regime. Relaxation is described reasonably well by a rate equation when lead-induced coherences are absent.

In the deep quantum regime, however, we find that the ISPI method is the only tool to obtain both the correct order of magnitude and the qualitative features of the relaxation rate as it depends on the system parameters $U$ and $J$. The same holds for the influence of $J$ and $U$ on the current in the deep quantum regime. Most importantly, the ISPI scheme proves to be useful in covering the full crossover regime where no small parameter can be identified and thus any perturbative approach becomes invalid.

Furthermore, the Kondo physics in such a single-spin system under non-equilibrium conditions is of course an interesting subject to study. It emerges when the Coulomb interaction is large compared to the tunnel coupling and the temperature is sufficiently low (also in comparison with the tunnel coupling). In this work, the two interaction terms in the Hamiltonian and the strong tunnel coupling at present limit the application of ISPI to intermediate temperatures. Therefore, Kondo features have not yet been obtained. In this regard, further development of the method is still demanding, see also the discussion in [48]. Nevertheless, due
to the suppression of long-time correlations at finite voltages, the regime of nonlinear transport where the Kondo correlations compete with the finite bias is accessible and will be treated in future work.

We have provided a first glimpse of the interesting new physics that is coming into reach with the ISPI scheme. A generalization of the model to several localized magnetic impurities, with electrons mediating a finite magnetization between them, should be possible. The real-time dynamics and all-electrical control of such devices could be simulated. The presented scheme is applicable for providing the $x$- and $y$-components of the impurity spin, thus yielding the complete spin dynamics and the real-time dephasing on the Bloch sphere.

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