Time-dependent transport through a correlated quantum dot with magnetic impurity

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Abstract. We investigate electronic- and spin transport through a single level quantum dot with magnetic impurity in a symmetric forward bias setup. On the quantum dot, electrons either interact with each other due to Coulomb interaction or with the spin 1/2 magnetic impurity. For certain configurations, the tunnel coupling to the leads induces an exponential relaxation of the impurity spin, which has been prepared in a polarized state initially. Furthermore, we study the influence of the nonequilibrium transport current on the relaxation dynamics. We obtain the respective numerical result by means of the iterative summation of path integral (ISPI) scheme. Within this approach, observables of interest are calculated from a functional derivative with respect to appropriate source terms in the Keldysh partition function. The real-time path integral extends over all possible paths (i) of the impurity spin and (ii) of the Ising like fluctuating spin fields we have to introduce in order to decouple the quartic interaction term of the Anderson model. The ISPI scheme allows us to sum up all paths including the time non-local self energies of the leads.

1. Introduction

Understanding nonequilibrium quantum transport in correlated nanoscale systems is presently in the focus of condensed matter research. Nanoscale quantum systems with an internal magnetic or spin structure such as nanomagnets, spin-doped quantum dots or single-electron transistors are the prime model systems for the complex physics involved. Those nonequilibrium conditions are induced by the coupling to a macroscopic (quantum or classical) environment. Recent physical questions that arise connect different research fields such as fundamental quantum mechanics, possible realizations of quantum computers and the design of novel (spin-)electronic devices [1, 2, 3].

In this article, we study the real-time quantum transport behaviour of a single-level quantum dot (SLQD) with an additional fixed magnetic spin-1/2 impurity on the dot. Besides the magnetic electron-impurity interaction, Coulomb correlations between dot electrons with mutual spins are taken into account. The ISPI [4] scheme allows us to study numerically the system under nonequilibrium conditions, when all appearing energy scales as Coulomb coupling, temperature, and magnetic interaction are of the same order of magnitude. We present results for the relaxation of the impurity spin and find an exponential decay as a function of time.
2. Model System

The model system consists of a SLQD, which is coupled to two metallic leads L and R via identical tunnelling barriers and a gate electrode to adjust the dot’s electrostatic potential $\Phi_{\text{dot}}$. Inside the SLQD resides a spatially fixed spin 1/2 magnetic impurity $M$, which interacts with the total spin of the dot electrons. Since the two-electron state $|d\rangle$ has to be a singlet with total spin zero, this electron-impurity interaction is only present for single occupation. The model allows for Coulomb repulsion in electron state $|d\rangle$ with scalar energy parameter $U \neq 0$ as well as a lifted degeneracy of electron $|\sigma\rangle$ and impurity (spin) states $|\tau\rangle$ with $\sigma, \tau = \pm 1 (= \uparrow, \downarrow)$. We describe the system via the Anderson-type Hamiltonian $\hat{H} = \hat{H}_D + \hat{H}_{\text{leads}} + \hat{H}_T + \hat{H}_{\text{INT}}$, with the individual parts $\hat{H}_D$ for the SLQD (without impurity), $\hat{H}_{\text{leads}} = \sum_p \hat{H}_p$ with $p = L, R (= \pm 1)$ representing the leads, the tunnelling/hybridization operator $\hat{H}_T$, and the electron-impurity interaction $\hat{H}_{\text{INT}}$. The dot-part without impurity is given by $\hat{H}_D = \sum_{\sigma} \epsilon d^\dagger_{\sigma} d_{\sigma} + U c^\dagger_{\uparrow} d_{\downarrow} d_{\downarrow}^\dagger$, where $d_{\sigma}$ is the creation operator for dot electrons with spin $\sigma$ and $\epsilon$ is the level energy. The leads are described as free electron gases with $\hat{H}_p = \sum_{kp\sigma} \epsilon_{kp} \hat{c}^\dagger_{kp\sigma} \hat{c}_{kp\sigma}$ and tunnelling between leads and dot by $\hat{H}_T = \sum_{kp\sigma} \gamma \hat{d}^\dagger_{\sigma} \hat{c}_{kp\sigma} + H.c.,$ where $\hat{c}_{kp\sigma}$ creates electrons in lead $p$ with spin $\sigma$, momentum $k$, and energy $\epsilon_{kp} = \epsilon_k - \mu_p$. The chemical potentials $\mu_p = pV_{\text{bias}}/2$ are tuned by a symmetrically applied bias voltage $V_{\text{bias}}$.

We model the spin-spin interaction between electron and impurity with Hamiltonian

$$\hat{H}_{\text{INT}} = J \sum_{\sigma = \pm 1} (\hat{\tau}_x \sigma d^\dagger_{\sigma} d_{\sigma} + \frac{\hat{\tau}_z \sigma}{2}),$$

(1)

where $J$ is the coupling strength. The longitudinal part (first term) of the interaction in (1) accounts for the shift of energy depending on the mutual spin alignment, whereas the transversal (second) term covers mutual spin flips of the impurity and electrons. The $\hat{\tau}_x$ with $x \in \{x,y,z,+,−\}$ denote the Pauli matrices for the impurity spin.

To calculate the non-equilibrium behaviour, we adapt the approach of the iterative summation of the path integral [4] to the present model. It is particularly suitable for systems with no small energy parameter and intermediate to long propagation times. We start by expressing the Keldysh partition function $Z$ (see [5]) as path integral over spin fields and fermionic Grassmann variables. To this end, we discretise time propagation along the Keldysh contour $C$ by introducing $N$ equidistant vertices each on the forward and backward branch. This defines a discretisation step $\delta_t$. Between two vertices on the forward branch, the system is propagated with time-evolution operator $\hat{U}_{\delta_t} = \exp\{-i\hat{H}_{\delta_t}\} = 1/2\sum_{s = \pm 1}\exp\{-i\hat{H}_{\delta_t}\}$ (operator $\hat{U}_{\delta_t}^\dagger$ on the backward branch). The Ising-like spin field $s$ is introduced during a discrete Hubbard-Stratonovich transformation [4], by which we map the Hamiltonian $\hat{H}$ of Coulomb interacting electrons to $\hat{H}^\prime$, which describes a system of free fermions coupled to $s$. $\hat{H}^\prime$ is obtained by replacing the quartic $\hat{H}_{\text{dot}}$ with the quadratic operator $\hat{H}_{\text{dot}} = \sum_{\sigma} (\epsilon^\prime_\sigma + i\lambda s) d^\dagger_{\sigma} d_{\sigma}$, where $\epsilon^\prime_\sigma = \epsilon_\sigma + U/2$ and $\lambda = \sinh^{-1}\sqrt{\sin(U\delta_t/2)}/i\sin^{-1}\sqrt{\sin(U\delta_t/2)}$.

At every vertex, unity operator partitions of coherent states for dot ($d$) and lead ($c$) electrons as well as for the discrete impurity spin basis ($1 = \sum_\tau |\tau\rangle\langle\tau|$) are inserted. Hence, in addition to the integration over Grassmann fields $c$ and $d$, the resulting path integral

$$Z = \sum_{\tau_1,\ldots,\tau_{2N}} \sum_{s_1,\ldots,s_{2N}} \int D[\alpha d\xi] (-1)^\ell \left(\frac{-i\Delta t}{2}\right)^{m} e^{iS[\alpha d\xi s]} \prod_{j \in T_{\text{up}}} d_{\tau_j}^\dagger d_{\tau_j} \prod_{k \in T_{\text{down}}} \xi_k^\dagger \xi_k$$

(2)

has to be summed over all tuple (paths) of the spin fields $\tau$ and $s$. Equation (2) is exact up to a normalization constant, which does not contribute to observables and therefore is omitted. For each impurity path, $m$ gives the number of flips (adjacent spins with opposite orientations), of
which \( \ell \) lie on the backwards contour. Since each term on the rhs of equation (2) is essentially an expectation value of a dot operator polynomial, only paths with even \( m \) (hence, an equal numbers of \( \overline{\sigma} \) and \( d_\sigma \) fields) contribute. The \( T_{\text{flip}}^\pm \) are sets of time indices (relating to real-time later vertex) of flips on the forward and backward branch in ascending order. The action reads

\[
S = \int_\mathcal{C} dt \sum_{\chi \in \{\sigma\}\{k\rho\sigma\}} \mathcal{T}_\chi(t) i\partial_t f_\chi(t) - \mathcal{H}[\overline{d}d\mathcal{E}\tau s] \quad f = c \text{ if } \chi \in \{k\rho\sigma\}, \text{ else } f = d, \tag{3}
\]

where \( \mathcal{H} \) is the result of replacing every operator in \( \hat{\mathcal{H}} \) by a corresponding time-dependent field. To get the expectation value of an operator \( \hat{O} \), we add the source term \(-\eta \mathcal{O}[\overline{d}d\mathcal{E}\tau s]\) to \( S \) and arrive at the generating function \( \mathcal{Z}[\eta] \). Again, the function \( O \) is obtained by substituting fields for operators. We can then write \( \langle \hat{O} \rangle = -i(\text{d} \ln \mathcal{Z}[\eta]/\text{d}\eta)|_{\eta=0} \). In the following, we are interested in the impurity orientation \( \langle \hat{\tau}_z \rangle \) but also the charge current \( \langle \hat{I} \rangle \) is implemented.

Since the integral over paths of Grassmann fields in \( \mathcal{Z}[\eta] \) is Gaussian, they can all be integrated out, if we assume that the leads are in thermal equilibrium. All calculations are done in the wide-band limit for the leads’ density of states \( \rho(\epsilon_{\text{lp}}) = \rho(\epsilon_{\text{fp}}) = \text{const.} \) with Fermi energy \( \epsilon_{\text{F}} \), that is absorbed in the coupling \( \Gamma := 2\pi\rho(\epsilon_{\text{F}})|\gamma|^2 \). In the final expression for the generating function

\[
\mathcal{Z}[\eta] = \sum_{\{r\},\{s\}} \prod_{\sigma} (-1)^\ell \left( \frac{J\delta\eta}{2} \right)^m e^{i\mathcal{S}_\mathcal{M}} \text{det} i\mathcal{G}_\sigma^{-1} \text{det} \Xi_\sigma, \tag{4}
\]

the determinants and the sum over paths \( \{r\} \) and \( \{s\} \) can be calculated iteratively, provided that not both \( V_{\text{bias}} \) and temperature \( T \) are zero or very small (see [4]). This is possible due to an exponential decay in real-time distance \( |t - t'| \) of the free leads’ Green’s function, which sets the dominant time-scale for dynamical correlations in the central region. Depending on the choice of observable \( \hat{O} = \hat{I}, \hat{\tau}_z \), the effective Green’s function \( \mathcal{G}_\sigma \) for spin \( \sigma \) dot electrons or the impurity action \( S_\mathcal{M} \) carries the \( \eta \)-dependence. The spin flip matrices \( \Xi_\sigma \) have dimension \( m/2 \) and are constructed from \( \mathcal{G}_\sigma \) as \( \Xi^{kl}_\sigma = \mathcal{G}_\sigma\{j_{2k},j_{2l-1}\} \) for \( \sigma = \tau_1, j_x \in T_{\text{flip}} = T_{\text{flip}}^+ \cup T_{\text{flip}}^- \), and \( \mathcal{G}_\sigma\{j,j'\} = \mathcal{G}_\sigma\{t_{j},t_{j'}\} \). For \( \sigma = -\tau_1 \), we have \( \Xi^{kl}_\sigma = \mathcal{G}_\sigma\{j_{2k-1},j_{2l}\} \).

3. Results

We studied the dynamics of the impurity spin orientation \( \langle \hat{\tau}_z \rangle \) in the presence of dot electrons, which are tunnelling on and off the dot and interact with the impurity. The initial state is set to be an empty dot with impurity wave function \( |\rangle \). Figure 1(a) shows the real-time behaviour of \( \langle \hat{\tau}_z \rangle \) for \( \epsilon = 0\Gamma, U = 0.5\Gamma, T = \Gamma, \text{ and } eV_{\text{bias}} = 0.6\Gamma \) and different values of the \( J \). It can be seen, that the impurity spin decays exponentially towards an unpolarized state with \( \langle \hat{\tau}_z \rangle(\infty) = 0 \). The decay time decreases with larger interaction strength \( J \). In figure 1(b), the influence of the bias voltage \( V_{\text{bias}} \) on the speed of this relaxation process is shown for \( J = \Gamma \) (other parameters as in 1(a)). Although we find a weak increase with growing values of \( V_{\text{bias}} \) and, consequently, increasing current through the dot (see inset in 1(b)), it is not a non-zero current that causes relaxation. Rather, it already occurs for \( V_{\text{bias}} = 0 \) due to the lead-dot coupling.

The exponential decay of \( \langle \hat{\tau}_z \rangle \) can be explained by a simple dynamical picture. As a result of the lead-dot hybridization, electrons tunnel stochastically on and off the dot. While a single electron is present, it interacts with the impurity via Hamiltonian (1), leading to mutual electron-impurity spin flips. The probability that a dot electron can induce flips during its stay on the dot increases with the rate \( J/\Gamma \). The value of \( J \) is connected to the number of flips per time unit, while \( \Gamma \) is proportional to the time between two tunnelling events and hence to the number of transitions between flipping single-electron and non-flipping two- or zero-electron states. For the chosen parameters, the stationary state of dot and impurity spins is unpolarized. Hence, electrons of both spin directions are present on the dot with equal probability and dynamically.
Figure 1. (a) Real-time relaxation of impurity spin orientation $\langle \hat{\tau}_z \rangle$ for different values of the electron-impurity interaction strength $J$ ($\epsilon = 0 \Gamma$, $U = 0.5 \Gamma$, $T = \Gamma$, and $eV_{\text{bias}} = 0.6 \Gamma$). In this configuration, a small unpolarized current flows through the dot. Since the impurity spin $M$ interacts with electrons of both spin directions with equal probability, the initial state with $\langle \hat{\tau}_z \rangle(0) = 1$ decays exponentially. All plot marks are numerical results, the solid lines are fitted curves with the decay time as single fit parameter. (b) Influence of the bias voltage $V_{\text{bias}}$ on the relaxation of the spin polarization. For all plots $J$ is equal to $\Gamma$ (all other parameters as in (a)). With increasing values of $V_{\text{bias}}$, the polarization decays faster. The inset shows the current $\langle \hat{I} \rangle$ for each value of $eV_{\text{bias}}$ with $I_0 = e\Gamma/h$.

decrease the spin polarization to zero. Growing values of $eV_{\text{bias}}$ between zero and $2\Gamma$ result in a rising current and a slight relative increase of the time ratio to find the dot in a spin flipping single-electron state, compared to the lower bias situation. This is the reason for the dependence of the spin relaxation time on $V_{\text{bias}}$ that is shown in figure 1(b).

4. Conclusions
We investigated the real-time dynamics of a SLQD with spin-1/2 magnetic impurity. An initially polarized impurity spin shows long-time transient behaviour, when coupled to metallic leads. The relaxation time mainly depends on the strength of the spin-spin interaction $J$ compared to the coupling $\Gamma$. Bias voltage and current have a considerably smaller impact on the relaxation time. In particular, even in the case of $V_{\text{bias}} = 0$, the polarization will decay due to electrons that tunnel stochastically between dot and leads. We provided a fully quantum mechanical description of this model system, which is of particular interest, since it has a sufficiently rich internal structure to study the correlated real-time dynamics of electron current and magnetic structure. Our exact method allows to tackle the most difficult and so far unexplored regime, where the Coulomb and the magnetic interaction are of comparable size and no small parameter exists. These results open the way for a direct and efficient control of the magnetization of a quantum dot by a nonequilibrium current.

References
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