

## Nonequilibrium Spin Dynamics in the Ferromagnetic Kondo Model

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Motivated by recent experiments on molecular quantum dots we investigate the relaxation of pure spin states when coupled to metallic leads. Under suitable conditions these systems are well described by a ferromagnetic Kondo model. Using two recently developed theoretical approaches, the time-dependent numerical renormalization group and an extended flow equation method, we calculate the real-time evolution of a Kondo spin into its partially screened steady state. We obtain exact analytical results which agree well with numerical implementations of both methods. Analytical expressions for the steady state magnetization and the dependence of the long-time relaxation on microscopic parameters are established. We find the long-time relaxation process to be much faster in the regime of anisotropic Kondo couplings. The steady state magnetization is found to deviate significantly from its thermal equilibrium value.

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**Introduction.**—Recently it has become experimentally feasible to trap isolated single molecules in nanogaps forming transistor geometries. In such molecular quantum dots a variety of interesting new phenomena have been observed. In case of a single C<sub>60</sub>-molecule attached to metallic leads (sketched in Fig. 1) the quantum phase transition between a singlet and a triplet eigenstate of the molecule has been studied in detail [1–3]. In particular, if the isolated molecule is prepared in the triplet configuration, its spin is partially screened by the conduction band. In this case the resulting effective exchange interaction between the residual spin and the conduction band is known to be ferromagnetic [4,5].

Replacing C<sub>60</sub> by a single-molecule magnet (SMM) such as Mn<sub>12</sub> gives rise to even more complex quantum impurity physics [6]. As a result of magnetic anisotropy induced by spin-orbit coupling, the large intrinsic spin of the SMM tends to align along the easy axis of the molecule. This gives rise to an energy barrier which suppresses magnetization reversal and makes SMMs promising candidates for applications such as high-density magnetic storage and quantum-information processing [7]. When coupled to metallic leads, the SMM can be described by an effective Kondo Hamiltonian with anisotropic exchange coupling between the impurity spin and the conduction band [8,9]

$$\mathcal{H} = \sum_{\vec{k}\sigma} \varepsilon_{\vec{k}} c_{\vec{k}\sigma}^\dagger c_{\vec{k}\sigma} + \frac{J_\perp}{2} \sum_{\vec{k}\vec{k}'} (c_{\vec{k}\uparrow}^\dagger c_{\vec{k}'\downarrow} S^- + c_{\vec{k}\downarrow}^\dagger c_{\vec{k}'\uparrow} S^+) + \frac{J_\parallel}{2} \sum_{\vec{k}\vec{k}'} (c_{\vec{k}\uparrow}^\dagger c_{\vec{k}'\uparrow} - c_{\vec{k}\downarrow}^\dagger c_{\vec{k}'\downarrow}) S_z + g\mu_B h S_z. \quad (1)$$

In the cotunneling regime it has been shown [10] that the exchange interaction is ferromagnetic, i.e.,  $J_\parallel \ll J_\perp < 0$ ,

if adding or subtracting an electron to the molecule increases the spin of the SMM. Preparing the system in a well-defined spin state and measuring the real-time spin dynamics can be achieved using electrical [11] or optical [12,13] field pulses, albeit experimental challenges in applying these techniques to molecular quantum dots still remain.

Although the Kondo Hamiltonian has been studied in great detail, the ferromagnetic regime has often been neglected (an exception is the investigation of the spatial equal-time spin-correlations of an underscreened spin-1 impurity [14]). In this Letter we will focus on two important questions arising in this context: By studying the magnetization dynamics we will investigate how fast an initially polarized spin will reduce its magnetization due to spin-flip scattering. For the antiferromagnetic Kondo model this question has been answered in [15–17]. Our analysis yields important information about the dominant relaxation mechanism in related experiments.

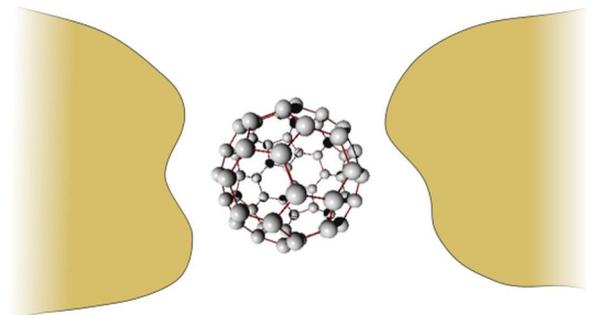


FIG. 1 (color online). Sketch of a C<sub>60</sub> molecule coupled to metallic leads.

One further important question regarding the relaxation process is the nature of the final state of the quantum system. Since a pure state remains pure under unitary time evolution, the complete system is not expected to behave like an equilibrium state even at long times. In equilibrium, the conduction electrons will not fully screen the spin, as is well known for the ferromagnetic Kondo model [18]. Instead, the coupling  $J_{\perp}$  provides weak spin-flip scattering which renormalizes the magnetization of the impurity spin to some finite value in the presence of a symmetry-breaking infinitesimal magnetic field. In the case of isotropic couplings and a spin  $S$ , this value is known as [18]

$$\langle S_z \rangle = S \left[ 1 + \frac{J\rho}{2} + O(J^2) \right], \quad (2)$$

where  $\rho$  is the density of states in the conduction band with support  $[-D, D]$  (we assumed  $\rho = (2D)^{-1}$  and employ units in which  $\hbar = k_B = D = 1$  in the following). However, the fact that the system's final state differs from thermal equilibrium does not necessarily imply that local observables retain a memory of the initial preparation, and in fact even for certain integrable systems the reduced density matrix of a local subsystem is known to thermalize [19]. For the ferromagnetic Kondo model, the low-energy spin-flip scattering rate renormalizes to zero, leaving open the question whether in the steady state at long times the impurity has finite magnetization or not. We will give a definite answer to this question in this letter and show that the asymptotic nonequilibrium magnetization at long times differs from the equilibrium value. Therefore in our model information about the initial preparation of the system is never completely lost, even for local observables.

*Methods.*—In recent years, several numerical approaches have been developed to calculate real-time dynamics of quantum impurity systems [17,20–23]. However, the accuracy of numerical data is usually not sufficient to give precise answers about the nature of the long-time decay, i.e., to identify analytical laws for the long-time tails and steady state values. We therefore use an analytical approach to identify the long-time behavior and compare it against numerical calculations to validate our analytical approximations. We first describe our analytical approach, before briefly sketching the numerical technique.

Within a poor man's scaling analysis a ferromagnetic exchange coupling of an impurity spin to a fermionic bath renormalizes to zero at the Fermi energy. This allows perturbative renormalization techniques to accurately describe the low-energy physics of such a system. In this context, a powerful technique is the flow equation method as invented by Wegner [24] and independently by Głazek and Wilson [25,26]. In a recent modification of the original flow equation method it has been shown that the underlying renormalization scheme can be extended to calculate the real-time evolution of interacting many-body systems [27–29]. As a notable feature, this approach allows to derive

exact analytical results. For further details of the flow equation approach and its application to the Kondo problem we refer to Ref. [30].

We briefly outline the main steps of the flow equation calculation. Details of this calculation will be published elsewhere [31]. As usual, the impurity spin operator is first transformed by a sequence of infinitesimal unitary transformations. The flowing spin operator has the form

$$S_z(B) = h(B)S_z + \sum_{kk'} \gamma_{k'k}(B): (S^+ s_{k'k}^- + S^- s_{k'k}^+);, \quad (3)$$

where the initial form of the operator is obtained for  $B = 0$  [30]. Here, the operators  $s_{k'k}^{\pm}$  are matrix elements of the conduction electron spin density raising and lowering operators. The coupling constants  $h(B)$  and  $\gamma_{k'k}(B)$  obey the flow equations

$$\begin{aligned} \frac{dh}{dB} &= \sum_{kk'} (\varepsilon_{k'} - \varepsilon_k) J_{k'k}^{\perp}(B) \gamma_{kk'}(B) n(k') [1 - n(k)] \\ \frac{d\gamma_{k'k}}{dB} &= h(B) (\varepsilon_{k'} - \varepsilon_k) J_{k'k}^{\perp}(B) + O(J^2), \end{aligned} \quad (4)$$

where  $n(k)$  denotes the Fermi distribution function. In addition, the flowing couplings  $J_{k'k}^{\perp}(B)$  and  $J_{k'k}^{\parallel}(B)$  of the Hamiltonian enter, which have to be calculated separately [30]. The fixed point of the transformation is reached in the limit  $B \rightarrow \infty$ , where we denote coupling constants in this basis by a tilde, e.g.,  $\tilde{h}$ . The Heisenberg equation of motion of the impurity spin can be solved efficiently by solving it first for the transformed impurity spin and reverting the unitary flow afterwards [27,28]. By solving the equations of motion in the diagonal basis of the Hamiltonian, one avoids secular terms that grow in an uncontrolled way with time and can obtain controlled analytical results even for the asymptotic long-time behavior.

In order to verify this semianalytical approach we employ the recently introduced time-dependent numerical renormalization group method (TD NRG) by Anders and Schiller [16]. Describing this method in detail is beyond the scope of this Letter. Let us only mention here that it is tailored to calculate the response of an (arbitrary) quantum impurity system to a sudden quench at time  $t = 0$  and is able to access the long-time-scales characteristic for Kondo physics. We refer the interested reader to Ref. [17] for more details on this method.

*Results.*—For our analytical calculations, we assume that the impurity spin is prepared in the up state of the spin projection operator  $S_z$  before the thermalized conduction electron bath ( $|FS\rangle$ ) is coupled to it, leading to a product initial state

$$|\psi\rangle = |\uparrow\rangle \otimes |FS\rangle. \quad (5)$$

At time  $t = 0$ , the spin is coupled to the conduction electrons, which would, e.g., be realized by attaching metallic leads to the single-molecule magnet. In the following, we restrict our calculations to spin  $S = 1/2$  and

zero magnetic field. Numerical calculations which we performed for the dynamics of larger spins  $S$  all satisfied the trivial relation  $\langle S_z(t) \rangle = S \langle \sigma_z(t) \rangle$ , which is exact up to  $O(J^2 S^2)$  from higher order flow equations [31]. Here  $S$  denotes the size of the spin and  $\sigma_z$  is the spin operator for spin  $S = 1/2$ . After solving the Heisenberg equation of motion for the operator  $S_z$ , the formal result for the magnetization reads

$$\langle S_z(t) \rangle = \frac{\tilde{h}}{2} + \sum_{kk'} \frac{\tilde{\gamma}_{kk'}^2}{2} \left( e^{it(\varepsilon_k - \varepsilon_{k'})} - \frac{1}{2} \right) n(k') [1 - n(k)]. \quad (6)$$

In the following, we will only discuss the purely quantum mechanical case, corresponding to  $T = 0$ . It turns out that the magnetization dynamics can be fully understood by the energy dependency of the couplings  $\tilde{\gamma}_{kk'}$ , which are obtained from a solution to Eq. (4).

*Isotropic Kondo Model.*—Let us first investigate an isotropically coupled spin. In equilibrium, perturbative scaling shows [32] that the isotropic coupling  $J$  logarithmically decreases upon reducing the half band width from  $D$  to some  $\Lambda < D$ ,

$$J(\Lambda) = \frac{J}{1 + \rho J \ln(\frac{\Lambda}{D})}. \quad (7)$$

At the low-energy fixed point, an infinitesimal magnetic field is sufficient to polarize the free spin, leading to a finite magnetization according to Eq. (2).

In nonequilibrium, our results show that the magnetization saturates as well, but to a different value than in equilibrium. Using the exact low-energy behavior of the couplings  $\tilde{\gamma}_{kk'}$  in Eq. (6), the asymptotic behavior of the magnetization is obtained as

$$\langle S_z(t) \rangle = \frac{1}{2} \left( \frac{1}{\ln(t) - \frac{1}{\rho J}} + 1 + \rho J + O(J^2) \right). \quad (8)$$

This behavior can be understood from the logarithmic renormalization of the coupling  $J$ , which directly enters the low-energy flow of the couplings  $\gamma_{kk'}$  via Eq. (4).

The steady state magnetization  $\langle S_z(t \rightarrow \infty) \rangle = \frac{1}{2} \times [1 + \rho J + O(J^2)]$  therefore differs from the equilibrium value as given by Eq. (2). The reduction from full polarization is  $\rho J/2$ , which is twice the equilibrium value. This can be attributed to the fact that the nonequilibrium dynamics starts with an impurity spin that is not dressed with a conduction band electron cloud: it therefore relaxes to a smaller value of the magnetization as compared to the dressed impurity spin in equilibrium.

A direct numerical solution of the flow equations allows to accurately determine the relaxation process also at intermediate and short time scales. Together with the analytical result from Eq. (8), this calculation can be compared to TD NRG calculations. Both methods yield very good agreement up to time scales of order  $t \approx 10^4$  where the asymptotic logarithmic relaxation is clearly visible; see Fig. 2. Increasing deviation of the curves for larger cou-

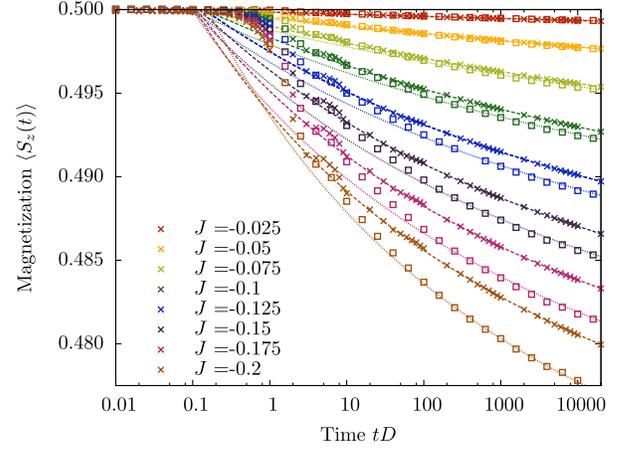


FIG. 2 (color online). Results for the *isotropic* ferromagnetic Kondo model ( $\times$ : TD NRG data,  $\square$ : flow equation data). Using our analytical result we fitted our data against  $\langle S_z(t) \rangle = (1 + aJ_{\parallel} + [\ln(t) - 1/(cJ_{\parallel})]^{-1})/2$  using  $a, c$  as fit parameters (lines).

pling strength  $J$  can be explained by the  $O(J^2)$  corrections to the flow equation result, which we neglected. We checked that the relative deviation of the two methods in terms of the quantity  $\langle S_z(t) \rangle - 1/2$  at some large but fixed time indeed grows approximately linearly in  $J$ . A fit of the TD NRG curves and the numerical implementation of the flow equation approach are in good agreement with the analytical result of Eq. (8).

*Anisotropic Kondo model.*—Studying the anisotropic Kondo model we restrict ourselves to the experimentally relevant case  $J_{\perp} > J_{\parallel}$  in the following. From a poor man's scaling analysis [32], it is known that the coupling  $J_{\perp}$  renormalizes as  $J_{\perp}(\Lambda) \propto \Lambda^{\rho} \sqrt{J_{\parallel}^2 - J_{\perp}^2}$  at low energies  $\Lambda$ . As in the isotropic case, the flow equation analysis shows that this behavior determines the asymptotic long-time relaxation of the spin, given explicitly by the power law

$$\langle S_z(t) \rangle = 0.5 \left[ 1 - \frac{\alpha^2}{2\tilde{g}_{\parallel}} t^{2\tilde{g}_{\parallel}} + \frac{\alpha^2}{2\tilde{g}_{\parallel}} + O(J^2) \right], \quad (9)$$

where  $\tilde{g}_{\parallel} = -\rho \sqrt{J_{\parallel}^2 - J_{\perp}^2}$ . The constant  $\alpha$  derives from the scaling equations for  $J_{\perp}$  and  $J_{\parallel}$ . Numerical checks show that it can be replaced by  $\alpha \approx \rho J_{\perp}$  as long as  $J_{\parallel} \lesssim 2J_{\perp}$ . In comparison to the isotropic case, the power-law decay of spin-flip scattering at low energies leads to much faster relaxation of the magnetization, whereas the steady state magnetization is enhanced. This behavior is reproduced by our numerical calculations shown in Fig. 3. Again, our calculations showed that the steady state magnetization  $\langle S_z(t \rightarrow \infty) \rangle = 1/2 + [\alpha^2/(4\tilde{g}_{\parallel})]$  is reduced twice as much from full polarization than in equilibrium. The analytical results are confirmed by numerical fits of our data; see Fig. 3. Let us point out that for the anisotropic Kondo model, our methods are starting from slightly different initial states. Using the flow equation approach one is restricted to a situation where the spin is initially com-

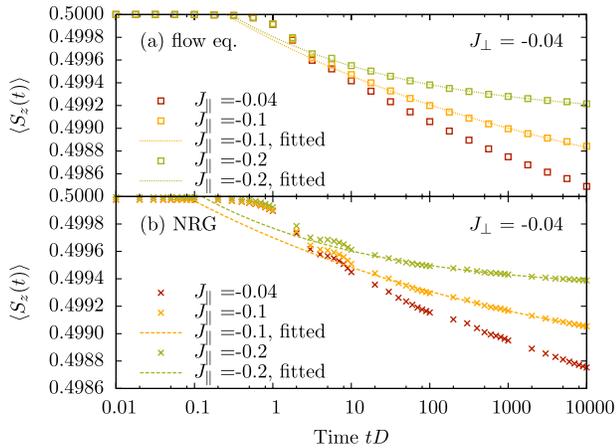


FIG. 3 (color online). For the *anisotropic* ferromagnetic Kondo model our numerical findings coincide with our analytical results using both methods. Fitting our data against  $\langle S_z(t) \rangle = at^{-\sqrt{J_{\parallel}^2 - J_{\perp}^2}} + c$  we found good agreement for the fit parameters  $a, c$ .

pletely decoupled from the fermionic bath. On the other hand, stability of the TD NRG algorithm in the anisotropic model requires preparing the polarized spin at time  $t < 0$  by applying a large magnetic field, while still allowing for a small exchange coupling to the metallic leads. The same long-time power-law relaxation was obtained with both methods. However, the slightly different initial states used in both methods become significant on short and intermediate time scales.

*Conclusions.*—We employed two different methods to analyze the real-time evolution of a ferromagnetically coupled Kondo spin, which is initially prepared in a polarized state. Exact analytical results for the long-time behavior of the magnetization were obtained for two different situations. For the isotropic ferromagnetic Kondo model, the long-time relaxation is logarithmic in time, whereas anisotropic couplings lead to a power-law decay at large times. Furthermore, exact analytical results for the asymptotic nonequilibrium magnetization were presented, which differ from the equilibrium magnetizations. They confirm that the local quantum impurity retains a memory of the initial preparation for asymptotically large times. This is due to the combined effect of nonequilibrium preparation and ergodicity breaking already in the equilibrium system.

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