Inelastic electron relaxation rates caused by spin-M/2 Kondo impurities

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We study a spin S = M/2 Kondo system coupled to electrons in an arbitrary nonequilibrium situation above Kondo temperature. Coupling to hot electrons leads to a shorter lifetime of pseudo particles and hence a larger Korringa width. This in turn is responsible for the enhanced inelastic relaxation rates of the electronic system. The rates are related to spin-spin correlation functions which are determined using a projection operator formalism. The results generalize recent findings for $S = \frac{1}{2}$ Kondo impurities which have been used to describe energy relaxation experiments in disordered mesoscopic wires.

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Recently, experimental evidence was found indicating that Kondo impurities might play an essential role for energy relaxation in mesoscopic gold wires¹ displaying much higher-energy relaxation rates than predicted by standard theory.² Based on these findings several theoretical studies have led to a qualitative or even quantitative explanation of experimental data by accounting for electron-electron interaction mediated by magnetic impurities.^{3–6} Assuming Kondo impurities of unknown origin as relevant inelastic-scattering centers also earlier experimental findings on copper wires⁷ could be explained.^{4–6} Moreover, assuming spin- $\frac{1}{2}$ impurities⁸ the detailed magnetic field dependence of energy relaxation experiments on copper wires⁹ could be fitted, strongly suggesting that Kondo impurities indeed play an essential role for energy relaxation at low temperatures.

In a recent work Anthore *et al.*¹⁰ studied energy relaxation in thin silver wires with Mn impurities and explained their findings using both, direct electron–electron interaction² and the effect of spin- $\frac{1}{2}$ impurities.⁸ Since Mn in silver is not a spin- $\frac{1}{2}$ impurity and furthermore the spin of the impurities in copper is not known, a generalization of the theory in Ref. 8 is desirable. In addition the impurity densities $c_{\rm imp}$ gained by fitting the energy relaxation data of the copper and silver samples typically exceed those obtained from measurements of the dephasing rate by more than an order of magnitude, see Refs. 8, 10 and papers cited therein. Impurity densities as high as those inferred from energy relaxation rates would lead to much higher dephasing rates than those found in experiments.

Considering the theoretical work in Refs. 3, 4, the impurity density can be lowered by increasing the spin *S* because only the product $S(S+1)c_{imp}$ enters the prefactor of the rate. However, this result does not take into account the spin dependence of the renormalized coupling constant. The aim of the present work is a generalization of the findings in Ref. 8 to arbitrary spin thereby exploring the possibilities of lowering the impurity density by increasing the spin *S*.

Since this work is an extension of Ref. 8 we follow the argumentation therein and, as far as possible, use the same notation. In order to make the paper self-contained, some of the basic ideas and definitions are, however, repeated. Whereas the technical details change, the main physical arguments remain the same and we refer the reader to Ref. 8 for further information.

We describe the quasiparticles and the impurity spin by the free Hamiltonian

$$H_0 = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} C_{\mathbf{k}\sigma}^{\dagger} C_{\mathbf{k}\sigma} - E_H S^z, \qquad (1)$$

where $C_{\mathbf{k}\sigma}^{\dagger}$ and $C_{\mathbf{k}\sigma}$ create and annihilate an electron in a given orbital **k**, and spin σ state. $\epsilon_{\mathbf{k}\sigma}$ is the energy of this state. The second term in Eq. (1) describes a spin M/2 impurity with Zeeman splitting $E_H = g \mu_B B$. The coupling between quasiparticles and impurity spin is described by the standard Kondo Hamiltonian

$$H_{I} = J_{0} \sum_{\mathbf{k}\mathbf{k}'\sigma\sigma'} \mathbf{S} \cdot \mathbf{s}_{\sigma'\sigma} C^{\dagger}_{\mathbf{k}'\sigma'} C_{\mathbf{k}\sigma}, \qquad (2)$$

where J_0 is the bare coupling and $\mathbf{s}_{\sigma'\sigma}$ denotes the vector of Pauli matrices. Here, we assume the impurity density $c_{\rm imp}$ to be small enough that we need to treat coupling to a single impurity only.

To determine the inelastic electron rates we consider the angularly averaged collision integral which in linear order in the density c_{imp} reads¹¹

$$I_{\sigma}(\boldsymbol{\epsilon}) = \frac{\iota}{\hbar} \{ f_{\sigma}(\boldsymbol{\epsilon}) \Sigma_{\sigma}^{>}(\boldsymbol{\epsilon}) + [1 - f_{\sigma}(\boldsymbol{\epsilon})] \Sigma_{\sigma}^{<}(\boldsymbol{\epsilon}) \}.$$
(3)

Here, $\Sigma_{\sigma}^{>/<}(\epsilon) = \Sigma^{>/<}(\mathbf{k}\sigma, \epsilon)$, where $\epsilon = \epsilon_{\mathbf{k}\sigma}$ is the electron self-energy on shell, assumed to be independent of the angular momentum. $f_{\sigma}(\epsilon)$ is the angularly averaged distribution function for electrons of energy ϵ and spin σ . For readability we suppressed the spatial dependence. Since the self-energy is proportional to the impurity density, we already replaced the electron Green's functions by their unperturbed form and integrated over frequency to get the classical form of the collision integral. In contrast to Ref. 8 we do not use the spin averaged self-energy but generalize the results to spin dependent distribution functions.

Our task is now to determine the electron self-energy which in turn leads to the electron scattering rates. Changing to the interaction picture and representing the spin degrees of freedom by pseudoparticles¹² one can use perturbation theory on the Keldysh contour to generate the graphs contributing to the electron self-energy. Since the topological structure of the graphs for a spin $\frac{1}{2}$ -system and a spin-M/2 system are the same, we can directly follow the reasoning in Ref. 8.

In lowest order the electron self-energy is given by a pseudofermion bubble and an electron or hole line in between. The pseudofermion bubble can be represented as a spin-spin correlation function which in frequency space directly determines the rates. Higher-order corrections are separable into terms adding an additional electron-hole pair and terms leading to higher-order corrections for a single electron-hole pair. The combinations of the second type are usually referred to as single particle intermediate state corrections and can be absorbed by a renormalization of the coupling constants.¹³ For arbitrary spin *S*, we find that the renormalized vertices J_{\pm}^{z} , J^{\pm} only depend on electronic occupation factors and therefore are given by relations very similar to those derived in Ref. 8. For a non-spin-flip processes we have

$$J_{\pm}^{z}(\epsilon)/J_{0} = \{ |1 - (\pi \rho J_{0})^{2} S(S+1)/4 - \rho J_{0}g_{\mp}(\epsilon \mp E_{H})|^{2} + (\pi \rho J_{0})^{2} S(S+1) \}^{-1/2}$$
(4)

and for a spin-flip process

$$J^{\pm}(\epsilon)/J_{0} = \{ |1 - (\pi \rho J_{0})^{2} S(S+1)/4 - \rho J_{0} [g_{\pm}(\epsilon) + g_{\mp}(\epsilon \pm E_{H})]/2|^{2} + (\pi \rho J_{0})^{2} S(S+1) \}^{-1/2}.$$
(5)

The renormalization is determined by the auxiliary function

$$g_{\pm}(\boldsymbol{\epsilon}) = \int_{-D}^{D} d\boldsymbol{\epsilon}' \frac{f_{\pm}(\boldsymbol{\epsilon}') - 1/2}{\boldsymbol{\epsilon} - \boldsymbol{\epsilon}' + i\delta}.$$
 (6)

In equilibrium this leads to the usual logarithmic corrections, however, the above formulas are applicable for arbitrary nonequilibrium situations. The Kondo temperature in this approximation reads

$$T_{K} = D \exp\left\{-\frac{1}{\rho J_{0}}\left[1 - \frac{(\pi \rho J_{0})^{2} S(S+1)}{4}\right]\right\}$$
(7)

and equals the bulk Kondo temperature. The phrase "above Kondo temperature" in this work means that the corrections determined by the auxiliary function (6) are still small compared to one. In this sense a system below the equilibrium Kondo temperature can be above Kondo temperature because of the nonequilibrium smearing of the distribution function.

Well above Kondo temperature it is usually assumed that all vertices renormalize independently. Therefore, one can equivalently put these renormalized quantities in a new interaction Hamiltonian

$$H_{I} = \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'} \{ S^{+}J^{+}(\boldsymbol{\epsilon}_{\mathbf{k}\uparrow}) C^{\dagger}_{\mathbf{k}'\downarrow} C_{\mathbf{k}\uparrow} + S^{-}J^{-}(\boldsymbol{\epsilon}_{\mathbf{k}\downarrow}) C^{\dagger}_{\mathbf{k}'\uparrow} C_{\mathbf{k}\downarrow} + S^{z} [J^{z}_{+}(\boldsymbol{\epsilon}_{\mathbf{k}\uparrow}) C^{\dagger}_{\mathbf{k}'\uparrow} C_{\mathbf{k}\uparrow} - J^{z}_{-}(\boldsymbol{\epsilon}_{\mathbf{k}\downarrow}) C^{\dagger}_{\mathbf{k}'\downarrow} C_{\mathbf{k}\downarrow}] \}, \qquad (8)$$

with energy and process dependent coupling constants. Using this Hamiltonian we have to restrict to elementary electron-hole pair excitations only. Other, more complex graphs of the one-particle intermediate state correction type are already put into the renormalization of the coupling constants. The electron self-energy is now given by the pseudofermion bubble coupled to arbitrarily many simple electronhole pairs with an electron or hole line in between and can be written as

$$\Sigma_{\sigma}^{>}(\boldsymbol{\epsilon}) = -i\sum_{\sigma'} \int d\boldsymbol{\epsilon}' W_{\sigma,\sigma'}(\boldsymbol{\epsilon},\boldsymbol{\epsilon}') [1 - f_{\sigma'}(\boldsymbol{\epsilon}')] \quad (9)$$

for the larger self-energy where $W_{\sigma,\sigma'}$ denotes the corresponding rates. The smaller self-energy $\Sigma_{\sigma}^{<}(\epsilon)$ is given by changing the variables (ϵ, σ) to (ϵ', σ') and $f \rightarrow 1-f$. Rewriting the pseudofermion bubble as spin-spin correlation function, the rates are given by

$$W_{-,+}(\epsilon,\epsilon') = \frac{c_{\rm imp}\rho}{4\hbar} J^{-}(\epsilon) J^{+}(\epsilon') C_{+}(\epsilon-\epsilon'), \quad (10)$$

$$W_{+,-}(\epsilon,\epsilon') = \frac{c_{\rm imp}\rho}{4\hbar} J^+(\epsilon) J^-(\epsilon') C_-(\epsilon-\epsilon'), \quad (11)$$

$$W_{+,+}(\boldsymbol{\epsilon},\boldsymbol{\epsilon}') = \frac{c_{\rm imp}\rho}{4\hbar} J_{+}^{z}(\boldsymbol{\epsilon}) J_{+}^{z}(\boldsymbol{\epsilon}') C_{z}(\boldsymbol{\epsilon}-\boldsymbol{\epsilon}'), \quad (12)$$

$$W_{-,-}(\epsilon,\epsilon') = \frac{c_{\rm imp}\rho}{4\hbar} J_{-}^{z}(\epsilon) J_{-}^{z}(\epsilon') C_{z}(\epsilon-\epsilon') \qquad (13)$$

with

$$C_{\pm}(t) = \langle S^{\pm}(t)S^{\mp}(0) \rangle, \quad C_{z}(t) = \langle S^{z}(t)S^{z}(0) \rangle.$$
(14)

Using Eq. (9) the collision integral takes the standard form for spin dependent scattering

$$I_{\sigma}(\boldsymbol{\epsilon}) = \sum_{\sigma'} \int d\boldsymbol{\epsilon}' \{ f_{\sigma}(\boldsymbol{\epsilon}) [1 - f_{\sigma'}(\boldsymbol{\epsilon}')] W_{\sigma,\sigma'}(\boldsymbol{\epsilon}, \boldsymbol{\epsilon}') - [1 - f_{\sigma}(\boldsymbol{\epsilon})] f_{\sigma'}(\boldsymbol{\epsilon}') W_{\sigma',\sigma}(\boldsymbol{\epsilon}', \boldsymbol{\epsilon}) \}.$$
(15)

The energies $\epsilon = \epsilon_{k\sigma}$ measure the kinetic energy and the Zeeman energy. Usually, when going over to a continuum description, the Zeeman splitting is put into a band bottom shift.

As in Ref. 8 we use a projection operator formalism to determine the correlation functions in an arbitrary nonequilibrium situation. Since the general formulas depend on the commutation relations of the spin operators only, we get the same expressions as in Ref. 8. The detailed formulas, however, change when evaluating impurity spin averages. Using the projection operators

$$P^{z}X = S^{z}\langle XS^{z}\rangle / \langle S^{z}S^{z}\rangle \quad \text{for } C_{z}$$
(16)

and

$$P^{\pm}X = S^{\pm}\langle XS^{\mp}\rangle / \langle S^{\pm}S^{\mp}\rangle \quad \text{for } C_{\pm}$$
(17)

one can derive a formally exact integrodifferential equation¹⁴

$$\dot{C}_{a}(t) = \Phi_{a}C_{a}(t) - \int_{0}^{t} du \,\phi_{a}(t-u)C_{a}(u)$$
(18)

with the solution in terms of the Laplace transform

$$\widetilde{C}_{a}(z) = \frac{C_{a}(t=0)}{z - \Phi_{a} + \widetilde{\phi}_{a}(z)}.$$
(19)

Here $a = z, \pm$ and, as in the $S = \frac{1}{2}$ case, $\Phi_z = \langle \dot{S}^z S^z \rangle / \langle S^z S^z \rangle = 0$, and $\Phi_{\pm} = \langle \dot{S}^{\pm} S^{\mp} \rangle / \langle S^{\pm} S^{\mp} \rangle = \mp i \tilde{E}_H$, which leads to the free propagation, where \tilde{E}_H includes the Knight shift neglected throughout this work. The averages are to be calculated self-consistently together with the steady-state electronic distribution functions f_{σ} and the occupation probabilities \mathcal{P}_m for the impurity spin being in state m.

The memory kernel $\phi_a(t)$ for the C_{\pm} correlation function reads

$$\phi_{\pm}(t) = \frac{\langle \dot{S}_{r}^{\pm}(t)\dot{S}^{\mp}\rangle}{\langle S^{\pm}S^{\mp}\rangle} + \Phi_{\pm}\frac{\langle \dot{S}_{r}^{\pm}(t)S^{\mp}\rangle}{\langle S^{\pm}S^{\mp}\rangle}.$$
 (20)

Here, the index r in $S_r^{\pm}(t)$ indicates that the dynamics of the spin operator is reduced by the projection. It is determined by the expression $\dot{S}_r^{\pm}(t) = \exp[i\hat{L}(1-P^{\pm})t]\dot{S}^{\pm}$ with the Liouville operator \hat{L} acting as $\hat{L}\hat{X} = [H,\hat{X}]/\hbar$. The memory kernel for the C_z correlation function is given by Eq. (20) with the replacements $\pm, \mp \rightarrow z$. We are interested in the regime well above Kondo temperature and expand the kernel up to second order in the renormalized coupling *J*. Since the dynamics of the expanded kernel function is oscillatory, the Fourier-transformed correlation function has always the simple form

$$C_{a}(\omega) = \frac{2C_{a}(t=0)\operatorname{Re}\phi_{a}(\omega)}{[\omega - i\Phi_{a} + \operatorname{Im}\phi_{a}(\omega)]^{2} + [\operatorname{Re}\phi_{a}(\omega)]^{2}} \quad (21)$$

with $a=z,\pm$. Further, we define $\operatorname{Re}\phi_a(\omega) \equiv \operatorname{Re}\{\tilde{\phi}_a(-i\omega + \delta)\}\)$ and the imaginary part $\operatorname{Im}\phi_a(\omega)$ follows from the Kramers-Kronig relation. When calculating the electronic distributions f_{σ} or spin occupation probabilities \mathcal{P}_m , the imaginary parts $\operatorname{Im}\phi_a(\omega)$ in the denominators lead to higher-order corrections in J and are neglected.

The damping rates (which were named $\nu_a(\omega)$ with $a = z, \pm$ in Ref. 8) read

Re
$$\phi_z(\omega) = \frac{\pi}{4} \sum_{\pm} \left[\frac{\langle S^{\pm} S^{\mp} \rangle}{\langle S^z S^z \rangle} \zeta_{\pm}(\omega \mp E_H) \right]$$
 (22)

for the C_z correlation function and

$$\operatorname{Re} \phi_{\pm}(\omega) = \frac{\pi}{4} \left[\zeta_{z}(\omega \mp E_{H}) + 4 \frac{\langle S^{z} S^{z} \rangle}{\langle S^{\pm} S^{\mp} \rangle} \zeta_{\mp}(\omega) \right] \quad (23)$$

for the C_{\pm} correlation functions. The auxiliary functions

$$\zeta_{z}(\omega) = \sum_{\pm} \int d\epsilon \rho^{2} J_{\pm}^{z}(\epsilon) J_{\pm}^{z}(\epsilon+\omega) f_{\pm}(\epsilon) [1 - f_{\pm}(\epsilon+\omega)]$$
(24)

and

$$\zeta_{\pm}(\omega) = \int d\epsilon \rho^2 J^{\mp}(\epsilon) J^{\pm}(\epsilon+\omega) f_{\mp}(\epsilon) [1 - f_{\pm}(\epsilon+\omega)]$$
(25)

describe coupling to electron-hole pairs. In equilibrium the damping leads directly to the Korringa width proportional to the temperature whereas in nonequilibrium this rate scales with a measure of the nonequilibrium situation, namely eU, leading to an increased inverse lifetime independent of the measurement temperature.

The equal-time correlation functions read for S = M/2

$$C_{z}(t=0) = \langle S^{z} S^{z} \rangle = \sum_{m=-M/2}^{M/2} \mathcal{P}_{m} m^{2}$$
(26)

and

$$C_{\pm}(t=0) = \langle S^{\pm}S^{\mp} \rangle = \sum_{m=-M/2}^{M/2} \mathcal{P}_{m}[S(S+1) - m(m\mp 1)].$$
(27)

As in the case $S = \frac{1}{2}$, the spin-spin correlation function $C(t) = \langle \mathbf{S}(t) \cdot \mathbf{S} \rangle = [C_+(t) + C_-(t)]/2 + C_z(t)$ fulfills the sum rule $C(t=0) = \int (d\omega/2\pi)C(\omega) = S(S+1)$ independent of the distribution \mathcal{P}_m .

To determine the master equation for the \mathcal{P}_m 's we use Eq. (27) and write the spin-flip correlation function as

$$C_{\pm}(\omega) \equiv \sum_{m} \mathcal{P}_{m}[S(S+1) - m(m \mp 1)]\check{C}_{\pm}(\omega). \quad (28)$$

The rate for the transition from state *m* to $m \pm 1$ then reads

$$\Gamma_{m \to m \pm 1} = [S(S+1) - m(m \pm 1)]\Gamma_{\pm}$$
⁽²⁹⁾

with

$$\Gamma_{\pm} = \frac{1}{4\hbar} \int d\omega \zeta_{\mp}(-\omega) \check{C}_{\mp}(\omega).$$
(30)

All other rates vanish. Note that the definition of Γ_{\pm} in this work is different from that employed in Ref. 8. The rate equations for the occupation probabilities

$$\dot{\mathcal{P}}_{m} = -\Gamma_{m \to m+1} \mathcal{P}_{m} - \Gamma_{m \to m-1} \mathcal{P}_{m} + \Gamma_{m+1 \to m} \mathcal{P}_{m+1} + \Gamma_{m-1 \to m} \mathcal{P}_{m-1}$$
(31)

with the normalization condition $\Sigma_m \mathcal{P}_m \equiv 1$ form a closed set of equations with the steady-state solution

$$\mathcal{P}_{m} = \frac{\Gamma_{+}^{M/2+m} \Gamma_{-}^{M/2-m}}{\sum_{n=0}^{M} \Gamma_{+}^{M-n} \Gamma_{-}^{n}}.$$
(32)

The probabilities obey the obvious balance relation $\mathcal{P}_m/\mathcal{P}_{m+1} = \Gamma_-/\Gamma_+$ which leads to the thermal distribution in equilibrium.

At vanishing magnetic field B=0 the probabilities are all equal, $\mathcal{P}_m = 1/(M+1)$, and the equal-time correlation func-

tions read $C_z(t=0)=S(S+1)/3$ and $C_{\pm}(t=0)=2C_z(t=0)$. If in addition the distribution functions are spin independent, the renormalized coupling constants become process independent $J_{\pm}^z = J^{\pm} \equiv J(\epsilon)$, and the auxiliary functions read $\zeta_z = 2\zeta_{\pm} \equiv 2\zeta(\omega)$. Inserting this in the correlation functions, we find $C(\omega) = [C_+(\omega) + C_-(\omega)]/2 + C_z(\omega) = 3C_+(\omega)/2$. In equilibrium and at low temperatures the width shrinks to zero and leads to $C(\omega) \rightarrow 2\pi S(S + 1)\delta(\omega)$.

The inelastic relaxation rate $1/\tau_{inel}$ at B=0 is the spinflip rate $1/\tau_{sf}$ reduced by the quasielastic rate, and we have $1/\tau_{inel} < 1/\tau_{sf}$. Quite generally, due to a sum rule for the spin-spin correlation function, the spin-flip rate obeys

$$\frac{1}{\tau_{\rm sf}} = \frac{1}{2} \sum_{\sigma,\sigma'} \int d\epsilon W_{\sigma,\sigma'}(\epsilon,\epsilon') = \frac{\pi}{2\hbar} \frac{c_{\rm imp}}{\rho} (\rho J)^2 S(S+1).$$
(33)

In order to discuss the possibility of reducing the impurity density by increasing the spin S at constant inelastic electronic rate, we may as well consider the spin-flip rate. As already explained in the Introduction, Eq. (33) suggests a decrease of the impurity density with increasing spin S. This is true only if the renormalization of the coupling constants is independent of S, i.e., at temperatures much higher than the Kondo temperature. To explain the experiments, however, ρJ has to be around 1/3 to be almost voltage independent. Otherwise the renormalization would not allow for the experimentally observed scaling property of the distribution function $f(\epsilon, eU) = f(\epsilon/eU)$, see Refs. 4, 8. In this regime, however, the renormalization depends on the spin S and scales for large spin like $\rho J \sim 1/\sqrt{\pi^2 S(S+1)}$ leading to a spin independent rate $1/\tau_{sf}$. Actually, the renormalized coupling constant equals the spin-flip t matrix⁴ which obeys a unitarity condition. It reaches a maximum, ρJ $=1/\sqrt{\pi^2 S(S+1)}$, at the Kondo temperature where the rate again would become independent of spin for all S. Although our theory is no longer valid in this regime, the outcome is quite physical since electrons always transfer the same spin when scattering from one impurity independent of *S*. This shows that using our theory an increase of the spin does not lower the impurity concentrations needed to describe the experiments. Even a more involved theory valid below Kondo temperature is not likely to help much since the scattering rate cannot exceed the limit discussed above.

To discuss the magnetic-field dependence of the rates W we consider two limiting cases. For low magnetic fields where the Zeeman splitting E_H is much smaller than the temperature or the applied voltage the occupation probabilities are all of the same order. Also the lifetimes do not change much and the behavior is dominated by the shift in the spin-flip correlation functions, $\omega \rightarrow \omega \pm E_H$. Therefore, there is no dependence on the spin *S* for small magnetic fields. For higher magnetic fields of the order of temperature or applied voltage, higher spin states are rapidly depopulated so that only two spin states like in the $S = \frac{1}{2}$ case lead to the dominant contribution. For higher *S* this is of course just a fraction and therefore in this regime the rates are even smaller than in the $S = \frac{1}{2}$ case.

In this work we have studied electron relaxation rates caused by magnetic impurities of arbitrary spin generalizing recent results for $S = \frac{1}{2}$. It is found that an increase of the spin *S* does not change the qualitative outcome and the rate at vanishing magnetic field is even unaffected by the spin for large *S*. Therefore, assuming magnetic impurities with higher spin *S* does not resolve the disagreement between Kondo impurity densities determined by energy relaxation experiments and weak localization experiments.

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