Oscillation and suppression of Kondo temperature by RKKY coupling in two-site Kondo systems

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Abstract. We apply our recently developed, selfconsistent renormalization group (RG) method [1] to STM spectra of a two-impurity Kondo system consisting of two cobalt atoms connected by a one-dimensional Cu chain on a Cu surface [2]. This RG method was developed to describe local spin screening in multi-impurity Kondo systems in presence of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Using the RKKY interaction of a one-dimensional chain, we explain the experimentally observed suppression and oscillation of the Kondo temperature, $T_K(y)$, as a function of the length of the chain and the corresponding RKKY interaction parameter $y$, regardless of the RKKY coupling being ferromagnetic or antiferromagnetic.

1. Introduction

The Kondo effect is a genuine many-body phenomenon which, in its usual magnetic manifestation, appears when a magnetic ion is immersed in a non-magnetic host metal. At high temperatures, the magnetic moment of the ion exhibits a Curie magnetic susceptibility corresponding to uncorrelated moments in a paramagnetic phase. When the temperature is decreased below a characteristic scale, the Kondo temperature $T_K$, the local moment is collectively screened by the conduction electron spins, leading to a narrow resonance in the electronic spectrum and a temperature-independent Pauli susceptibility [3, 4]. The Kondo effect is observed in a variety of systems, ranging from metallic alloys [5, 6] to artificial semiconductor nanostructures like quantum dots [7-10] and molecular devices [11-13].

In a spin-1/2 two-impurity Kondo (2IK) system, two different ground states are possible, the Kondo state where each of the two local spins forms a Kondo singlet with the conduction electrons, and a dimer state where the two local spins are mutually bound into a singlet or triplet. The two states are distinguishable by different behavior of the spin correlation functions between a local spin and the conduction electrons on one hand and between the local spins among each other on the other hand. It has been recognized [1] that it makes a crucial difference whether there is a direct exchange coupling between the impurities or whether the interimpurity coupling is mediated by the conduction electrons, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [14, 15, 16, 17]. With a direct exchange coupling, a quantum phase transition between the Kondo and the dimer phase [18, 19] occurs only for a particular particle-hole symmetry [20], associated with a coupling-decoupling transition of the conduction band. In the case of RKKY-mediated interimpurity coupling, a complete decoupling of the conduction electrons at
The two-impurity Kondo system is described by the Hamiltonian,

\[ H_{\text{2IK}} = \sum_{k \sigma} \varepsilon_{k \sigma} c_{k \sigma}^\dagger c_{k \sigma} + J_0 \sum_{i=1,2} \mathbf{S}(x_i) \cdot \mathbf{S}(x_i), \]

where \( \mathbf{S}(x_i) \) and \( \frac{1}{2} \mathbf{S}(x_i) \) represent the spin operators of the impurities and conduction electrons at the positions of the impurities, \( x_{1,2} \), respectively. \( (k, \sigma) \) represents a complete basis of the conduction electron system, composed of the substrate and the 1D Cu chain. Density functional theory (DFT) calculations indicate \[2\] that the RKKY coupling is dominated by the 1D chain (because in the 3D substrate the RKKY interaction decays faster than in 1D) and that the local DOS in the Cu chain is well approximated by an infinitely long chain. Deviations from this behavior are found only for the shortest chains of length \( n = 1 \), see section \[3\]. We, therefore, approximate the conduction-mediated coupling between the Co spins at positions \( x_1 \) and \( x_2 \) by the RKKY coupling of two spins separated by \( x = x_2 - x_1 \) on an infinite chain (infinite chain approximation). In a simple model for the Cu chain we assume a Fermi momentum of \( k_F = \pi/(2a) \) corresponding to half filling of the Cu 4s band, where \( a \) is the Cu lattice spacing.

In leading order perturbation theory in the local spin exchange coupling \( J_0 \), the RKKY coupling reads, \( J_{\text{RKKY}}(x) = -\chi_c(x) \), where \( \chi_c(x) \) is the free conduction electron susceptibility (or polarization) of the chain \[30\], as shown in Fig. 1 (a). We calculate \( \chi_c(x) \) here directly in position space, as this will be needed for the RG vertex in section \[3\] and is more concise than the momentum space calculation \[30\].

\[ \chi_c^R(x_2, x_1; \Omega) = \int_{-\infty}^{+\infty} d\varepsilon \ n_F(-\varepsilon) \left( G_c^R(x; \varepsilon + \Omega) A_c(-x; \varepsilon) + A_c(x; \varepsilon) G_c^A(-x; \varepsilon + \Omega) \right), \]

where \( G_c^{R/A} \) is the retarded/advanced Green’s function for the free electrons and \( A_c \) represents their spectral function. \( n_F(\varepsilon) \) denotes the Fermi-Dirac distribution, and \( \varepsilon = E - \varepsilon_F \) is the energy with respect to the Fermi level, \( \varepsilon_F \). The static susceptibility entering the RG vertex \[1\] is obtained from the zero-frequency limit \( \Omega \to 0 \), and \( n_F(\varepsilon) = \Theta(\varepsilon) \) for temperature \( T = 0 \).
Assuming, for simplicity, a quadratic dispersion, $\varepsilon_k = k^2/2m$, the 1D free Green’s function (per spin orientation) reads

$$G_0^{0,R/A}(x, \omega) = \mp i\pi N(\omega) \exp[\pm ik(\omega)|x|] ,$$

where $N(\omega) = m/(2\pi k(\omega))$ is the continuum conduction electron DOS and $k(\omega) = \sqrt{2m(\varepsilon_F + \omega)}$ the momentum corresponding to the energy $\omega$. Hence, the static conduction electron susceptibility is real and reads,

$$\chi_c^R(x_2, x_1; \Omega = 0) \equiv \chi_c^R(\rho; \Omega = 0) = -\frac{1}{2} N(0) \left( \text{Si}(\rho) - \frac{\pi}{2} \right) ,$$

where $\rho = 2k_F|x|$, and $\text{Si}(z) = \int_0^z dx \frac{\sin(x)}{x}$ the integral sine. Note that the long-distance limit of $\chi_c^R(x; \Omega = 0)$ is

$$\chi_c^R(x; \Omega = 0) \overset{|x| \to \infty}{\sim} \frac{\cos(\rho)}{\rho} ,$$

which manifests its slow decay and oscillatory behavior as a function of interimpurity spacing.

3. Renormalization group analysis for symmetrically coupled impurities

In the previous section, we calculated the RKKY interaction between the two Co impurities mediated by the 1D Cu chain connecting the magnetic ions. The RKKY interaction implies that the total spin exchange vertex between conduction electrons and a local impurity spin acquires nonlocal contributions: A conduction electron can scatter from impurity 2, and that spin flip can be transferred to impurity 1 by the itinerant conduction electrons travelling along the Cu chain from the 2nd to the 1st Co impurity. This process is depicted diagrammatically in Fig. 1 (b). As seen from the figure, this transfer process necessarily involves the total, local spin susceptibility $\chi_f$ of the 2nd impurity. Its exact behavior is known from the Bethe ansatz solution [32]. At low temperatures, $T < T_K$, it is proportional to $1/T_K$ and crosses over to Curie behavior $\chi_f \sim 1/T$ for $T \gg T_K$. With the expression for the RKKY-modified Kondo vertex, the RG equations for the Kondo coupling can be obtained by the same procedure as developed in Ref. [1]. The RG equation
for the dimensionless Kondo coupling, \( g = gN(0) \), as a function of the running conduction bandwidth, \( D \), is obtained as \([1]\),

\[
\frac{dg}{d\ln D} = -2g^2 \left( 1 - y g_0^2 \frac{D_0}{T_K} \frac{1}{\sqrt{1 + (D/T_K)^2}} \right),
\]

(6)

where \( D_0 \) is the full bandwidth of the conduction electrons, \( g_0 = J_0 N(0) \) the dimensionless, bare Kondo coupling, \( T_K = T_K(y) \) is the RKKY-modified Kondo scale (to be determined), and \( y \) is a dimensionless RKKY strength parameter. It is obtained for a 2IK system as (c.f. Ref. \([1]\)),

\[
y = \frac{8W}{\pi^2} \text{Im} \left[ \frac{e^{-ikFX}}{N(0)^2} G_c^R(x, \Omega = 0) \chi_c(g, \Omega = 0) \right] = \frac{8W}{\pi N(0)} \cos(g) \chi_c(g, \Omega = 0),
\]

(7)

with \( W = 2 \) the Wilson ratio of a single Kondo impurity. Note that the term proportional to \( \frac{1}{T_K} \) on the right-hand side of Eq. (6) originates from the behavior of the local impurity susceptibility, \( \chi_f \propto \frac{1}{T_K} \) \([2]\). Furthermore, note that the RKKY parameter \( y \) of the 2IK system is generically positive, even though \( \chi_c^R(x, 0) \) has an alternating sign, as shown in Fig. 2 left panel. Technically, this is because in \( y \) the oscillatory factors \( \exp(-ikFX) G_c^R(x, \Omega = 0) \) and \( \chi_c(g, \Omega = 0) \) combine to form an essentially positive term. It is physically expected because the interimpurity RKKY coupling should reduce the onsite Kondo spin fluctuations irrespective of the RKKY coupling being ferro- or antiferromagnetic.

The RG equation (6) is straight-forwardly integrated by separation of variables,

\[
\int_{g(D_0)}^{g(D)} \frac{dg}{g_2^2} = \int_{D_0}^{D} \frac{dD}{D} \left( 1 - y g_0^2 \frac{D_0}{T_K} \frac{1}{\sqrt{1 + (D/T_K)^2}} \right).
\]

(8)

To deduce the Kondo temperature \( T_K \), one should use the fact that, as the running cutoff reaches the Kondo scale \( D \to T_K \), the effective exchange coupling diverges, \( g \to \infty \), indicating that the RG flows to the strong-coupling fixed point with complete Kondo screening. Thus, the Kondo scale, defined in this way, is generally finite even though \( g \to \infty \). By equivalence of both Kondo sites, it is the same as the Kondo temperature \( T_K \) appearing in the local spin susceptibility \( \chi_f \) on the second Kondo site (see above). This yields the defining equation for \( T_K \) \([1]\),

\[
\ln \left( \frac{T_K(y)}{T_K(0)} \right) = -y \alpha g_0^2 \frac{D_0}{T_K(y)},
\]

(9)

where \( D_0/T_K(0) \gg 1 \) was used and the coefficient \( \alpha = \ln(\sqrt{2} + 1) \) arises from fixing the integration constant by the condition \( D(g \to \infty) = T_K(y) \). Note that this is an implicit or selfconsistent equation, because the \( \beta \)-function itself [right-hand side of Eq. (6)] depends parametrically on \( T_K \) via \( \chi_f \). The solution of Eq. (9) also indicates that in the 2IK system the Kondo scale is modified by the RKKY interaction \([1]\), i.e., \( T_K = T_K(y) \). In the absence of the RKKY interaction, the bare Kondo coupling, \( g_0 \equiv g(D_0) \), satisfies

\[
\frac{1}{2g_0} = \ln \left( \frac{T_K(0)}{D_0} \right).
\]

(10)

Hence, by defining a rescaled Kondo temperature, \( \tau(y) := \frac{T_K(y)}{T_K(0)} \), one obtains the equation for the RKKY-modified Kondo scale as

\[
\tau(y) = e^{-y \alpha g_0^2 \frac{D_0}{T_K(0)}} \tau(y).
\]

(11)
Figure 2: Left panel: The universal curve for the RKKY-modified Kondo temperature, $T_K(y)/T_K(0) = \frac{-y/(e^{y/e_c})}{W(-y/(e^{y/e_c}))}$, as a function of the normalized RKKY strength parameter, $y/y_c$, where the critical RKKY parameter for Kondo breakdown is $y_c = 1/(e^{\gamma_0})$ and $e \approx 2.718$ is Euler’s constant (see Eq. (12)). Complete Kondo singlet formation ceases to exist for $y > y_c$. Right panel: Oscillation and suppression of the Kondo temperature, $T_K(y(n))/T_K(0)$, for Cu chains of length $n$: Comparison of theory and experiment Ref. [2]. In the experiment, the single-ion Kondo scale is $T_K(0) = 110$ K. The overall amplitude of $T_K(y(n))/T_K(0)$ is obtained by scaling the theoretical value to the experimental one for $n = 2$. It is remarkable that the 1D infinite chain model accurately reproduces the experimental data even for short chains of $n \geq 2$. The RKKY coupling, $I_{RKKY}(n)$ (red solid line) and the RKKY strength parameter, $y(n)$, averaged over one lattice spacing $a$ (dashed green line), are also shown (arbitrary units).

It can be solved in terms of the Lambert $W$ function, $^1$

$$\tau(y) = \frac{-\gamma_0 y}{W(-\gamma_0 y)},$$

with the parameter,

$$\gamma_0 = g_0^2 \frac{D_0}{T_K(0)} = \frac{\alpha}{4(ln(T_K(0)/D_0))^2} \frac{D_0}{T_K(0)}.$$  

The solution $\tau(y)$ is shown in Fig. 2 left panel [1]. It is seen that the solution of Eq. (12) ceases to exist when $y$ exceeds the critical strength, $y > y_c = 1/(e^{\gamma_0})$, indicating that the RG flow does not diverge, i.e. a strong coupling Kondo singlet is not formed for $y > y_c$. As seen from Eq. (13), the critical strength $y_c$ is determined alone by the single-impurity Kondo scale $T_K(0)$. At the breakdown point, the RKKY-induced suppression ratio of the Kondo scale takes the universal, finite value $T_K(y_c)/T_K(0) = 1/e \approx 0.368$, where $e$ is Euler’s constant [1]. We emphasize that, since the solution $1/e \leq T_K(y)/T_K(0) \leq 1$ for all values of $1 \geq y \geq 0$, the term in brackets on the right-hand side of the RG equation (10) ($\beta$-function) remains always positive and less than one, i.e., the RG flow remains always in the perturbatively controlled regime.

$^1$ Indeed, the first branch of the Lambert function, $W_0$, is the solution. More information can be found in Ref. [33].
4. Comparison with experimental results and discussion

Having adapted the RG formalism of Ref. [1] to the Kondo suppression in a 2IK system RKKY-coupled by a 1D Cu chain, we are now in a position to compare with the corresponding experiments of Ref. [2]. All spatial dependence is cast in the distance dependence of the RKKY parameter $y(n)$, Eq. (7). Since the magnetic moment of a Co impurity atom is spread over a Co $d$-orbital occupying about one unit cell, we average $y(n)$ over one lattice spacing $a$ of the underlying Cu lattice (green dashed curve in Fig. 2 right panel) and insert the result into Eq. (12) to calculate the RKKY-induced Kondo suppression ratio $T_K(y(n))/T_K(0)$ as a function of the Cu chain length. Since the RKKY wave number is fixed to $2k_F = \pi/a$ (half band filling, see section 2), this result has a single scale factor, $\gamma_0$, as the only adjustable parameter to fit the experimental results. We adjust it to fit the experimental data for $n = 2$. Note that, in fact, the relative suppression, $T_K(y(n))/T_K(0)$, with respect to the single-ion value of experimentally $T_K(0) = 110$ K [2] has no adjustable parameter at all. As seen in Fig. 2 (right panel), the agreement with the experimental data for $n \geq 2$ is quantitatively very good, with only a slight deviation for a Cu chain length of $n = 6$. It is remarkable that the 1D infinite chain model reproduces, without fit parameter, the periodicity as well as the relative amplitude of the $T_K(y(n))$ suppression accurately even for short chain lengths down to $n = 2$, considering the crudeness of this model. It is important to note that our theory explains that the RKKY interaction of either sign (ferro- or antiferromagnetic) suppresses the Kondo scale, as is physically expected and experimentally observed [2]. Only for the shortest Cu chain, $n = 1$, the theory does not fit the experiment. We attribute this to the fact that the simple infinite-chain model fails for this very short cluster, because the local DOS and, hence, the RKKY interaction are strongly influenced by boundary effects, the geometry of the wave functions and chemical bonds. More precise DFT calculations should be employed to calculate the RKKY coupling and the $y$-parameter in this case. Interestingly, however, the strongest experimentally observed suppression ratio is $T_K(y(n = 1))/T_K(0) = 46/110 \approx 0.42$ (Fig. 2 right panel), which is remarkably close to the theoretical prediction $1/e$, considering that the impurity spacing cannot be changed continuously in the experiment.

In summary, we have given a short review of the recently developed renormalization group method to tackle the influence of the RKKY interaction on the Kondo screening in multi-impurity and lattice Kondo systems [1]. The application of this theory to the oscillation and suppression of the Kondo scale in Co-Cu$_n$-Co chains [2] yield generally good, quantitative agreement. This further supports the validity of this RG method. The deviation of the theory from the experiment for the shortest chain, $n = 1$, is attributed to boundary effects not included in the infinite chain model and may be improved by DFT calculations of the input parameters of the RG.

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