Overscreened Kondo physics in four-fold symmetric Au systems doped with Co

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Abstract. We analyze the relation between a model recently proposed to describe Co doped Au chains with four-fold symmetric leads, and the generalized SU(2)⁴SU(2) Anderson model. We compare the structure of the Kondo limit in the general case. We calculate the spectral density and the conductance as a function of temperature in both cases, showing that both models display similar non-Fermi liquid behavior.

1. Introduction

The Kondo effect is one of the paradigms in modern solid state theory. In its simplest version, the spin 1/2 of a magnetic impurity is screened by the spin of a conduction band (also called conduction channel) below a characteristic temperature $T_K$, resulting in a singlet ground state [1]. In nanoscience, ideal systems with a single localized spin 1/2 were studied and confirmed predictions of the SU(2) Kondo impurity model, or the more general SU(2) impurity Anderson model [2, 3, 4]. More recently, nanoscopic systems were studied [5, 6] in which a spin 1 was underscreened by one conduction channel, leaving a doublet ground state, and having a quantum phase transition to a singlet ground state [6], as predicted in an Anderson model which mixes a singlet and a triplet with a doublet through one conduction channel [7].

The physical realization of nanostructures with overscreened spins is more difficult, due to the sensibility of the physical properties to asymmetry of the channels or inter-channel charge transfer [8] which spoil the observation of the expected non-Fermi liquid behavior [8, 9, 10]. Recently, it has been shown that if in Co doped Au chains, the axial symmetry along the chain is broken by a sufficiently strong crystal field of four-fold symmetry, which splits the 3$d$ $x^2−y^2$ and $xy$ levels of Co, then the system displays clear signatures of non-Fermi liquid behavior, like an impurity contribution to the entropy $\ln(2)/2$ and a conductance per channel at low $T$ of the form $G(T) \simeq G_0/2 − a\sqrt{T}$, where $G_0$ is the conductance at zero temperature in the one-channel case [11]. A similar behavior is expected for Co doped Cu chains attached to a four-fold symmetric lead [12].

A model studied before which displays non-Fermi liquid behavior is the SU(N)×SU(M) generalization of the Anderson model [13, 14], where M identical conduction channels screen the N degrees of freedom of the impurity. It has been shown that for the multichannel non-Fermi-liquid case $M \geq N$, the non-crossing approximation (NCA) reproduces the exact power-law
behavior of the main physical properties, including the spectral density of states [13, 14].

In this work we show that the model mentioned above for Co doped nanoscopic systems, can be considered as a generalization of the SU(2) × SU(2) Anderson model. We also calculate the spectral density and the conductance for both models using the NCA. We show that rescaling the Kondo temperature, the low-temperature properties of both models become very similar.

2. Model
First principles calculations [11] show that the ground state of the Co ion is a quadruplet of the 3d orbital configuration with the xz and yz orbitals occupied by only one electron, and also either the xy or x² − y² orbital is singly occupied, depending on the (irrelevant) sign of the crystal field. We denote the components of this quadruplet with three holes in the 3d shell by |M3⟩ where M3 is the spin projection. This configuration is hybridized via the hopping to the xz and yz 5d orbitals of Au with two 3d⁸ triplets (|αM3⟩) in which either the xz or the yz orbital of Co is doubly occupied. The effective Hamiltonian which describes this physics is [11]

\[
H = \sum_{M3} \left( E_3 + \frac{D}{2} M_3^2 \right) |M3⟩⟨M3| + (E_2 + D'M_2^2) \sum_{αM2} |αM2⟩⟨αM2| + \sum_{νkσ} \epsilon_{νkσ} c_{νkσ}^† c_{νkσ} \\
+ \sum_{αM2M3νkσ} V_ν(\frac{1}{2} M_2σ |3\frac{1}{2} M3⟩⟨αM2⟩⟨M3| + H.c.),
\]

where \(c_{νkσ}^†\) creates a hole in the 5d band of Au states with symmetry α at the left (ν = L) or right (ν = R) of the Co site. A crucial role is played by the splitting \(D'\) between the quadruplet states with spin projection ±3/2 and those with \(M_3 = ±1/2\). Instead, \(D'\) which splits the excited states with \(M_2 = ±1\) from those with \(M_2 = 0\), has a negligible effect for realistic values [11]. Both terms appear as a consequence of the spin-orbit coupling \(H_{SOC} = λ \sum_i \hat{1}_i \cdot \hat{s}_i\). In spite of the irrelevance of \(D'\) for the system studied, it is interesting to note that for \(D, D' → +∞\), the model becomes equivalent to the SU(2) × SU(2) Anderson model with a hybridization \(V_ν\) reduced by a factor \((2/3)^{1/2}\), which is the value of the Clebsch-Gordan coefficients \((1\frac{1}{2}0\frac{3}{2}σ)\) in Eq. (1). This is easily seen using two pseudofermions to represent the states |M3⟩ with \(M_3 = ±1/2\) and two pseudobosons for |0⟩. In the general case we need four pseudofermions and six pseudobosons to represent all states for the NCA treatment. The formalism is similar to that used by two of us for the singlet-triplet Anderson model [15].

Further insight into this equivalence is given by a Schrieffer-Wolff canonical transformation that eliminates the hybridization term (the last one) in Eq. (1) for infinite \(D\) (actually \(D \gg T_K\) is enough, where \(T_K\) is the Kondo temperature [11]), and leads to an effective exchange interaction between an effective spin 1/2 (\(S\)) for the states |M3⟩ with \(M_3 = ±1/2\) and the spin of the extended states at the impurity site (\(\hat{s}^α\)). The resulting Kondo model has the form

\[
\hat{H}_{2CK} = \frac{2}{3} V_L^2 + V_R^2 \sum_α \left( \frac{2D' + δ}{D' + δ} \hat{S}_z^α \hat{s}_z^α + 2\hat{S}_x^α \hat{s}_x^α + 2\hat{S}_y^α \hat{s}_y^α \right) + \sum_{νkσ} \epsilon_{νkσ} c_{νkσ}^† c_{νkσ},
\]

where \(δ = E_2 - E_3 - D/8\) is the charge-transfer energy (we set the Fermi energy \(ε_F = 0\)). For \(D' → +∞\), the interaction is the usual one for the isotropic two-channel Kondo model (2CKM), except for the prefactor 2/3 which comes from the above mentioned Clebsch-Gordan coefficients. For \(D' = 0\) the coefficient of the \(\hat{S}_z^α \hat{s}_z^α\) term is exactly half the other two, and the model reduces to an anisotropic 2CKM, which also has non-Fermi liquid properties.
3. Effect of spin-orbit coupling

To calculate $D$ we have solved exactly the Hamiltonian of the 3$d^7$ configuration [16] including $\hat{H}_{SOC}$. We show in Fig. 1 how $D$ depends on the splitting $\Delta E$ between 3$d$ $x^2−y^2$ and $xy$ orbitals on $D$. We took the values of the energies of the different 3$d$ orbitals from $ab\text{ initio}$ results [11]: $E_{3z^2−r^2} = −0.1\ \text{eV}$, $E_{xz} = E_{yz} = 0.6\ \text{eV}$, and assume $−E_{x^2−y^2} = E_{xy} = \Delta E/2$. For the Coulomb integrals $F_i$ [16] and $\lambda$, we have taken the values $F_2 = 0.16\ \text{eV}$, $F_4 = 0.011\ \text{eV}$, $\lambda = 0.08\ \text{eV}$, obtained from a fit of the low energy spectra of late transition metal atoms. From the figure, one sees that for $\Delta E = 0$, $D = −0.0426\ \text{eV}$ is negative (80% of the value $−2\lambda/3$ expected from first order perturbation theory in $\hat{H}_{SOC}$) indicating that the Co spin is oriented along the chain. As $\Delta E$ increases, $D$ also increases and changes sign for $\Delta E \approx 0.8\ \text{eV}$. For the value $\Delta E = 1.2\ \text{eV}$ estimated from $ab\text{ initio}$ calculations when a lead with four-fold symmetry is attached to the Co atom [11], we obtain $D = 0.0017\ \text{eV}$.

Stretching the device, the interatomic positions increase, reducing the effect of the crystal field and therefore decreasing $D$. This renders in principle possible to tune the system from the overscreened regime which takes place for $D > 0$ to the non Kondo regime for $D < 0$ through the underscreened regime for $D = 0$.

4. Spectral density and conductance

Figure 1. Splitting between states with $S_z = ±3/2$ and $S_z = ±1/2$ of the isolated Co atom in a Au chain, as a function of the splitting between Co $xy$ and $x^2−y^2$ orbitals

Figure 2. Spectral density per channel and spin for $D, D' \to +\infty$ (full line) and $D = 0.002, D' = 0$. Other parameters are $W = 1$, $\delta = 0.67$, $\Delta = 0.225$, and $T = 0.005T_K$.

Figure 3. Conductance as a function of temperature for the same parameters as in Fig. 2. $G(0) = 1.2G_0$, where $G_0 = 2e^2/h$. 
For the NCA calculations, we have taken the half band width of the conduction band \( W = 1 \) as the unit of energy and the resonant level width \( \Delta = \pi \rho_0 (V_D^2 + V_R^2) = 0.225 \), where \( \rho_0 = 1/(2W) \) is the density of states of the free conduction band. In Fig. 2 we compare the spectral density \( \rho_{\alpha\sigma}(\omega) \) of the SU(2)\( \times \)SU(2) Anderson model with that for more realistic values of the anisotropies. The main difference is that in the latter case, the Kondo temperature \( T_K \) defined from the half width at half maximum of the Kondo peak, decreases by a factor near 4, due to the decrease of the \( z \) component of the effective exchange interaction [see Eq. (2)] and two shoulders appear near energies \( \pm D \) due to the effect of the \( |M_3\rangle \) states with \( M_3 = \pm 3/2 \), which are neglected in the SU(2)\( \times \)SU(2) model. For \( D, D' \to +\infty \), we obtain \( T_K = 2 \times 10^{-4} \), while for \( D = 0.002, D' = 0, T_K = 5 \times 10^{-5} \). Note that in contrast to Fermi liquid systems, for which \( \pi \rho_{\alpha\sigma}(0)\Delta = 1 \) in the Kondo limit, here the correct value is 1/2. The NCA gives a value slightly larger.

In Fig. 3 we show the dependence of the conductance \( G(T) \) on temperature. As before, the value \( G(0) = 1.2G_0 \) is nearly half the value \( 2G_0 \) expected for a Fermi liquid with two channels. The deviation of 20\% is due to shortcomings of the NCA. Moreover, the \( \sqrt{T} \) dependence that \( G(T) \) shows for \( T < 0.2T_K \) is also characteristic of non-Fermi liquid behavior. The slope is very similar for both sets of parameters when it is scaled by the corresponding \( T_K \).

5. Discussion

We have shown that the model Eq. (1) contains the SU(2)\( \times \)SU(2) Anderson model as a special case. Using the NCA we show that the non-Fermi liquid structure of the former is kept for more realistic parameters. Since the NCA can be extended to calculate the conductance out of equilibrium, this opens the possibility to study to what extent non-Fermi liquid behavior can be seen for a finite applied bias voltages \( V_b \). In principle one expects that \( G(V_b) \) has a dependence of the type \( \sqrt{V_b} \) for \( eV_b \) smaller than a fraction of \( k_BT_K \).

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