Relevance of quantum fluctuations in the Anderson–Kondo model

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Abstract. We study a localized spin coupled to an Anderson impurity to model the situation found in higher transition metal or rare earth compounds like e.g. LaMnO$_3$ or Gd monopnictides. We find that, even for large quantum numbers of the localized spin, quantum fluctuations play an essential role for the case of ferromagnetic coupling between the spin and the impurity levels. For antiferromagnetic coupling, a description in terms of a classical spin is appropriate.

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1. Introduction

Transition metal oxides show a fascinating complex behaviour in their electronic properties [1]. This complexity stems from the interplay between the formation of narrow 3d-bands leading to a delocalization of these states on the one hand and the local part of the Coulomb interaction between the 3d-electrons tending to localize them [1]. While compounds of the early 3d elements like e.g. LaTiO$_3$, which typically accommodate one 3d electron, can at least qualitatively be described in terms of a one-band Hubbard model [2]–[4], materials involving higher transition metal elements like LaMnO$_3$ or TlSr$_2$CoO$_5$ require the use of a model including the full 3d shell. In particular, to understand the magnetic properties and the frequently occurring metal-insulator transitions [1] one has to take into account the interplay between density- (‘Hubbard $U$’) and exchange-type (‘Hund’s $J$’) contributions to the local Coulomb interaction. Note that similar features can also be found in compounds involving higher rare earth elements, for example the rare earth pnictides.

A particularly interesting example is La$_{1-x}$Ca$_x$MnO$_3$ [5, 6]. Besides is complicated phase diagram comprising a large variety of paramagnetic and magnetically ordered metallic and insulating phases one finds a colossal magneto-resistance (CMR) [7]. In this cubic perovskite the five-fold degenerate 3d level is split by crystal field into three-fold degenerate $t_{2g}$, which have the lower energy, and two-fold degenerate $e_g$ states. These states have to be filled with $4 - x$ electrons, nominally yielding a metal even for $x = 0$. However, taking into account the local Coulomb interaction, three of these electrons will occupy the $t_{2g}$-states forming an $S = 3/2$ high-spin state due to Hund’s coupling, which interacts ferromagnetically with the electron occupying the $e_g$ states. Ignoring the Coulomb repulsion among the electrons in the $e_g$ subsystem, one encounters the well-known double-exchange model [8], which has been extensively studied as suitable model for manganites (see e.g. references in [9]). In most of these investigations, however, the $t_{2g}$-spin was approximated by a classical moment to allow the use of standard techniques like e.g. quantum Monte Carlo (QMC) [10]–[16]. A particular drawback of such an approximation is that the results are independent of the sign of the exchange coupling to the localized spin [9]. However, without such a replacement, one is typically restricted to low-order diagrammatic techniques ([17] and references therein).

A more realistic treatment should of course also include the local Coulomb interaction within the $e_g$ subsystem. Such a model has been proposed recently [9] and studied in the framework of the dynamical mean-field theory (DMFT) [18]. Again, the $t_{2g}$-spin had to be replaced by a classical moment to allow the solution of the DMFT equations with QMC.

In this paper, we want to study the validity of approximating the quantum spin by a classical object. To this end, we investigate the simplest possible model, namely a quantum impurity model consisting of a local orbital with interacting charge degrees of freedom coupled to a non-interacting host and a spin.

While such a model surely cannot access every aspect of the physics of the corresponding lattice model, it is the basic ingredient in a DMFT calculation and thus understanding its fundamental properties is of importance to properly interpret results obtained in a DMFT calculation. Moreover, although such a calculation will focus on local dynamics only, one can obtain at least qualitative results about possible ordered phases, too [18]. To this end it is viable to obtain a feeling of how the additional spin will modify local charge and spin properties.
We employ Wilson’s numerical renormalization group (NRG) \([19, 20]\) to solve this model. This technique allows us to treat the model in the whole parameter regime and in particular identify small energy scales if present.

The paper is organized as follows. In the next section we present the model and briefly review its properties for a classical spin in the limit of vanishing Coulomb interaction. The presentation of our results follows in section 3. The paper closes with a summary and discussion.

2. The model

The simplest model that allows us to obtain an idea how the coupling to an additional local spin-degree of freedom influences the properties of correlated electrons is

\[
H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{\sigma} \left( \epsilon_d + \frac{U}{2} d_{-\sigma}^\dagger d_{-\sigma} \right) d_{\sigma}^\dagger d_{\sigma} + \frac{V}{\sqrt{N}} \sum_{k\sigma} (c_{k\sigma}^\dagger d_{k\sigma} + \text{h.c.}) - J_K \sum_{\alpha, \beta} \vec{S} \cdot \vec{s}_d. \tag{1}
\]

The first three terms represent a conventional single-impurity Anderson model (SIAM) \([21, 22]\), while the last contribution introduces an additional spin degree of freedom which couples to the local states via an exchange interaction. Similar models have been studied with various techniques in connection with double quantum dots \([23]–[28]\). However, in these cases the additional impurity was represented by a correlated charge degree of freedom coupled via a hopping. In the limit of half-filling and weak interimpurity hopping, this system maps to our model with \(S = 1/2\) and vanishing antiferromagnetic \(J_K\). This limit shows interesting physics on its own \([27, 29]\), for example two-stage Kondo screening or quantum phase transitions. However, these models neither allow for ferromagnetic \(J_K\), large and possibly anisotropic \(J_K\) nor spins \(S > 1/2\), which are the particular cases we are interested in here.

Of course our model (1) does not fully represent the situation found in e.g. LaMnO\(_3\) since it lacks the orbital degrees of freedom. On the other hand, NRG calculations for multi-orbital models are extremely expensive \([30]\) and we believe that as far as the qualitative aspects are concerned this simplification will not substantially modify the validity of our observations for the more complicated model.

For a classical spin \(\vec{S}\), the model (1) can be solved exactly for \(U = 0\) \([31, 32]\). The result for the single-particle Green function of the \(d\) states is

\[
G_d(z) = \frac{1}{2} \left( \frac{1}{z - \epsilon_d + i\Delta_0 + J_K} + \frac{1}{z - \epsilon_d + i\Delta_0 - J_K} \right), \tag{2}
\]

where \(\Delta_0 = \pi N_F V^2\) and we assumed \(|\vec{S}|^2 = 1\) and a flat conduction density of states (DOS) of infinite width and value \(N_F\). The resultant DOS \(\rho_d(\omega) = -\frac{1}{\pi} \text{Im} G_d(\omega + i\delta)\) shows two peaks of width \(\Delta_0\) centred at \(\epsilon_d \pm J_K\). Note that one can view this result as spin-averaged DOS of the SIAM at \(U = 0\) in an external magnetic field of strength \(J_K\) and that the result is independent of the sign of \(J_K\).
3. Results

3.1. Classical limit

Neither for a quantum spin nor for a classical spin and $U > 0$ an exact solution exists. However, it is tempting to extend the above interpretation for a classical spin in the following way: solve a standard SIAM in an external magnetic field of strength $J_K$ and average over the resulting spectra for spin up and down. Note that this procedure again leads to results that do not depend on the sign of $J_K$.

Since the additional spin enters only on the local level, we use the standard NRG algorithm [20, 33, 34] to solve the impurity problem and calculate physical quantities. To obtain reliable spectra in a magnetic field, we furthermore employ the technique proposed by Hofstetter [35]. Since a discretization of the energy axis introduced in the NRG leads to discrete spectra, a broadening must be introduced to obtain smooth results for dynamical quantities. Again, we follow the standard procedure here [33].

As example we present in figure 1 calculations for the SIAM in a magnetic field, averaged over the spin direction, together with results for the SIAM with additional Ising spin and large quantum spin $S = 10$ (‘classical spin’). The parameters for the SIAM are $U/\pi \Delta_0 = 5.3$ and $\epsilon_d = -U/2$. The NRG discretization parameter was $\Lambda = 2.5$ and we kept 1000...4000 states per iteration depending on the size of the local spin. The NRG-spectra finally were broadened with a Gaussian of width $b = 0.6$ [33, 36].

The black curve in figure 1 was obtained from a calculation with a magnetic field $B = 8 \times 10^{-3} \Delta_0$, the red curve with a local Ising spin $S = 1/2$ and coupling $J_K = 4B$, the blue and magenta curves with local spin $S = 10$ and couplings $J_K = \pm 2B/10$ to simulate the classical limit $S \rightarrow \infty$. The values of $J_K$ were scaled such that $B = J_K s_d S$. The inset shows...
Figure 2. Entropy and effective squared impurity moment for local spin $S = 1/2$ and different values of $J_K$ as function of $T/T_K(0)$, where $T_K(0)$ is the Kondo temperature of the model with $S = 0$. Full (dashed) lines denote antiferromagnetic (ferromagnetic) coupling. The other impurity parameters are $U/\Delta_0 = 6.4\pi$ and $\epsilon_d = -U/2$.

In the region around $\omega = 0$ the calculation with Ising spin and magnetic field coincide perfectly, yielding a splitting of the original Kondo resonance at $B = J_K = 0$, as expected. However, the ‘classical limit’ with $S = 10$ differs considerably. Depending on the sign of $J_K$, either a Kondo resonance (ferromagnetic coupling) or a gap (antiferromagnetic case) emerges at $\omega = 0$ in addition to the splitting of the original Kondo peak. Apparently, even for such a large value of $S$ the effect of quantum fluctuations is still prominent. However, the corresponding energy scales are considerably reduced compared to $J_K = 0$ and we expect the results to converge to the anticipated one as $S \to \infty$.

3.2. Quantum spins: $S = 1/2$

Let us now turn to the discussion of the effects of a local quantum spin on the low-energy properties. In figure 2 the impurity contribution to the entropy (upper panel) and the effective impurity moment $\mu_{\text{eff}}^2 := T \cdot \chi_{\text{imp}}$ (lower panel) for different values of $J_K$ for a local spin $S = 1/2$ are shown as function of $T/T_K(0)$, where $T_K(0)$ denotes the Kondo scale for the system with $S = 0$. The SIAM parameters were $U = 6.4\pi\Delta_0$ and $\epsilon_d = -U/2$. The NRG discretization, number
of states kept, etc were chosen as for figure 1. The full lines in figure 2 represent results for antiferromagnetic coupling $J_K$, the dashed lines those for ferromagnetic coupling.

Decreasing $J_K$ from $J_K = 0$ to some antiferromagnetic $|J_K| < T_K^{(0)}$ results in the scenario depicted by the full red curves in figure 2. Around the temperature $T_K^{(0)}$ screening occurs as in the normal Kondo effect, resulting in a situation with effective moment $\mu_{\text{eff}}^{(0)} = 1/4$ and entropy $\ln 2$ that resembles a free spin $S = 1/2$ again. For a much lower $T_K^{(1)}$, a second screening takes place to the ground state with $S = 0$. This second screening can be interpreted in the following way. At $T < T_K^{(0)}$ the Kondo screening of the local level of the SIAM by the conduction states leads to the formation of a local Fermi liquid [22]. This local Fermi liquid is characterized by an effective mass $m^* \propto \rho_{QP}(0) \propto 1/T_K^{(0)}$ [22], where $\rho_{QP}(0)$ denotes the quasi-particle DOS at the Fermi level of the local Fermi liquid. Thus, for temperatures $T \ll T_K^{(0)}$ we are left with an effective Kondo model, with antiferromagnetic exchange coupling $J_K$. The Kondo scale of this effective low-temperature model is then given by $T_K^{(1)} \propto \exp(-1/(2\rho_{QP}(0)|J_K|)) \propto \exp(-\alpha T_K^{(0)}/|J_K|)$. This effect has been observed before by several authors [27]–[29] and baptized two-stage Kondo screening. When $J_K$ becomes of the order of $T_K^{(0)}$, the Kondo screening is replaced by the formation of a local singlet with an energy scale $\approx |J_K|$.

For ferromagnetic coupling $J_K > 0$, on the other hand, the proper fixed point is—as in the conventional Kondo model—the local-moment one with residual entropy $\ln 2$ and effective moment $1/2$. Again, from the dashed curves in figure 2 one can distinguish two regimes. For $J_K < T_K^{(0)}$ we observe screening on the scale of $T_K^{(0)}$, the additional local spin effectively behaving like a free spin all the way down to $T = 0$. However, for $J_K \gg T_K^{(0)}$ the impurity first forms a local spin triplet (entropy $\ln 3$ and moment 2/3). This moment is then partially screened at a reduced Kondo scale $T_K \ll T_K^{(0)}$; energy scales and physical properties will behave as in the conventional underscreened Kondo model [37, 38].

The behaviour discussed previously is reflected in the local DOS depicted in figure 3. As already noted in the classical limit, the coupling to the additional spin leads to a corresponding shift of the upper Hubbard band, which however is for larger $|J_K|$ more pronounced for antiferromagnetic exchange. In this case one also clearly sees the two-stage screening at $|J_K| < T_K^{(1)}$ and the formation of the local singlet at $|J_K| > T_K^{(0)}$ (inset to upper panel of figure 3), suppressing the Kondo screening. Here, one always finds a gap in the DOS at $\omega = 0$, which size is set by $T_K$. For ferromagnetic coupling, on the other hand, the inset in the lower panel of figure 3 proves that the screening resonance remains intact, but shows a width decreasing with increasing $J_K$.

### 3.3. Quantum spins: general $S$

How does the behaviour discussed in the previous section change with increasing spin quantum number $S$ or more precisely, do we recover a ‘classical’ result for large enough $S$? Let us start with a discussion of the weak-coupling results shown in figure 4. The calculations were done in the weak-coupling regime $U/\Delta_0 = 1$ for fixed value $S(S+1)/\Delta_0 = 1/3$ to achieve the same classical energy scale for all $S$. We did calculations up to $S = 10$, which, according to our results in subsection 3.1, we expect to be already very close to the classical limit. Indeed, for $S = 10$ (green curves in figure 4) we do find almost identical behaviour for $J_K < 0$ and $J_K > 0$ except for extremely low temperatures. Note however, that even for this large value for $S$ the DOS for $\omega/\Delta_0 < 0.1$ for $J_K < 0$ does not reach the full unitary limit due to quantum fluctuations.
Figure 3. Local DOS for an impurity spin $S = 1/2$ and different values of $J_K$ as a function of $\omega/\Delta_0$. Impurity parameters as in figure 2. The upper panel collects the results for antiferromagnetic coupling, the lower for ferromagnetic. The insets show the spectra for $\omega > 0$ in a semi-logarithmic plot. For antiferromagnetic $J_K$ one here clearly sees the two-stage Kondo effect for $|J_K| < T_K^{(0)}$, i.e. a formation of an Abrikosov–Suhl resonance at $T_K^{(0)}$ and the formation of a singlet at $T_K^{(1)}$ (cf also discussion of figure 2).

Figure 4. Local DOS for fixed $\sqrt{S(S+1)}|J_K|/\Delta_0 = 1/3$ as a function of $\omega/\Delta_0$ in the weak-coupling regime $U/\Delta_0 = 1$ for different values of local spin $S$. The upper panel collects the results for antiferromagnetic coupling, the lower panel those for ferromagnetic. Note that for $J_K > 0$ the influence of the local spin is very small, i.e. the curves are nearly indistinguishable. The insets show the spectra for $\omega > 0$ in a semi-logarithmic plot.
The differences are more dramatic for small values of $S$. As expected, for $J_K < 0$ there occurs a Kondo screening with an energy scale $T_K(S)$ decreasing exponentially with increasing $S$. Note that even for comparatively large $S = 3/2$ the influence of quantum fluctuations is still pronounced and appears in an energy regime that may still be of experimental relevance.

The differences become even more pronounced if we increase the local Coulomb repulsion to $U/\Delta_0 = 10$, which lies in the intermediate-coupling regime with a Kondo scale $T_K/S_0 \approx 0.07$. Since thus $J_K > T_K/S_0$ we do not expect two-stage screening here. The results for otherwise same model parameters are collected in figure 5. Note that due to the choice of $J_K$ the Hubbard bands do not move with increasing $S$. Furthermore, as can best be seen from the insets to figure 5, in all cases $S > 0$ we observe additional shoulders, respectively peaks, in the DOS at $\omega \approx \pm \sqrt{S(S+1)}|J_K|$. The low-energy behaviour, however, is markedly different for $J_K < 0$ (upper panel in figure 5) and $J_K > 0$ (lower panel in figure 5). Below $\omega/\Delta_0 < 0.07 \approx T_K^{(0)}$, the former case always develops a (pseudo-) gap due to the formation of a singlet between the local degrees of freedom, while the latter tends to recover a Kondo-resonance for the total spin, again with exponentially decreasing $T_K(S)$. Owing to the universal behaviour of the conventional Anderson model for $U/\pi \Delta_0 > 1$ (‘Kondo regime’) we actually expect the behaviour observed here to be generic in this parameter regime. Quite obviously, while in the weak coupling regime the ‘classical limit’ is reached already for moderate values of $S$, one has to be very careful when dealing with the strongly correlated regime.

4. Summary and conclusion

In this paper we presented calculations for an extended Anderson impurity model, where the local charge degrees of freedom in addition couple to a localized spin. The motivation to study such a
model is based on the observation that in a variety of transition metal or rare earth compounds, the complex local orbital structure can be split into a localized spin, which can take large values $S \gg 1/2$, coupled via Hund’s exchange to a more delocalized set of possibly also correlated states. A particular example surely is the famous LaMnO$_3$.

The solution of this model for different regimes of model parameters was accomplished by using Wilson’s NRG, which provides accurate and reliable results for thermodynamics and dynamics and is able to resolve arbitrarily small energy-scales that may appear in the problem. The findings can be summarized as follows: even for comparatively large localized spin $S = 10$, we still observe the influence of quantum fluctuations on the properties of the impurity charge degrees of freedom. These effects become more pronounced when these charge degrees of freedom are correlated themselves as to be expected for example in LaMnO$_3$. Note, however, that the temperature scales related to these quantum effects will be small compared to e.g. the typical ordering temperatures in LaMnO$_3$, but can possibly be more relevant in rare earth compounds. Depending on the ratio $|J_K|/T_K^{(0)}$, where $T_K^{(0)}$ is the Kondo temperature for the model without additional spin, different regimes can be identified, which in contrast to the classical prediction do markedly depend on the sign of $J_K$.

Thus, for the solution of correlated lattice models with such an additional spin degree of freedom within DMFT one has to be likely careful when using the approximation of a classical spin, even when $S$ is comparatively large. Moreover, the expected physics can be read off our results right away, at least for half-filling. Due to the reduced Kondo scale for Hund’s type or ferrromagnetic coupling, we expect a corresponding reduction of a critical $U$ for a Mott–Hubbard transition. On the other hand, for antiferromagnetic exchange coupling the corresponding $U_c = 0$ at $T = 0$, because the forming of a local singlet immediately leads to an insulating state.

Quite interesting are also the magnetic properties of the system. Again, we may anticipate from the impurity calculations that antiferromagnetism is still the prevailing magnetic order, but in the vicinity of certain commensurate band fillings we can also expect ferromagnetic order from a corresponding RKKY exchange. These investigations are currently in progress.

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