Strong-coupling scenario of a metamagnetic transition

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We investigate the periodic Anderson model in the presence of an external magnetic field, using dynamical mean-field theory in combination with the modified perturbation theory. A metamagnetic transition is observed which exhibits a massive change in the electronic properties. These are discussed in terms of the quasiparticle weight and densities of states. The results are compared with the experimental results of the metamagnetic transition in CeRu2Si2.

By “metamagnetic transition”, we describe an anomalous behavior of the magnetization as function of external field \( b_{\text{ext}} \), namely a sudden increase at a finite field \( b_{\text{ext}} \). Many rare-earth materials exhibit this kind of behavior. One has to distinguish between those materials which already show long-range (e.g. antiferromagnetic) order for zero-field and those which are paramagnetic.

In the first case, the materials already have finite local moments at zero-field. Here a metamagnetic transition occurs when the external field is stronger than the internal (antiferromagnetic) exchange between these moments. This situation is found for example in CeFe-based alloys.

The other possibility, where a paramagnetic substance enters a high-magnetization state at a critical field \( b_{\text{crit}} \), is realized by some heavy-fermion metals as for example CeRu2Si2 (PAM) with an external magnetic field. The periodic Anderson model (PAM) with an external magnetic field \( H_{\text{ext}} \) couples equally to the spin of the localized electrons with a band of uncorrelated conduction electrons. These two electronic sub-systems are coupled by a hybridization term. The Hamiltonian of the PAM reads:

\[
H = \sum_{\vec{k}, \sigma} \epsilon(\vec{k}) n_{\vec{k}\sigma}^\dagger n_{\vec{k}\sigma} + \sum_{i, \sigma} \epsilon_i f_{i\sigma}^\dagger f_{i\sigma} + V \sum_{i, \sigma} (f_{i\sigma}^\dagger n_{i\sigma} + s_{i\sigma}^\dagger f_{i\sigma}) + \frac{U}{2} \sum_{i, \sigma} n_{i\sigma}(f_{i\sigma}^\dagger)^2,
\]

(1)

Here, \( n_{\vec{k}\sigma} \) and \( n_{i\sigma} \) are the creation and annihilation operators for a conduction electron with Bloch vector \( \vec{k} \) and spin \( \sigma \) (a localized electron on site \( i \) and spin \( \sigma \)) and \( n_{i\sigma}(f_{i\sigma}^\dagger) = f_{i\sigma}^\dagger f_{i\sigma} \) (an on-site Coulomb interaction strength between two \( f \)-electrons). Throughout this paper, the conduction band will be described by a free (Bloch) density of states, \( \rho_0(E) = \frac{1}{N} \sum_{\epsilon} \delta(E - \epsilon(\vec{k})) \), of semi-elliptic shape. Its width \( W = 1 \) sets the energy scale, and its center of gravity the energy-zero: \( T_{ii} = \frac{1}{N} \sum_{\epsilon} \epsilon(\vec{k}) = 0 \). The magnetic field \( b_{\text{ext}} \) is given in energy units of the band width \( W \) (\( z_\sigma = +1(-1) \) for \( \sigma = \uparrow(\downarrow) \)). The external field couples equally to the spin of the \( f \) and conduction band electrons. This is in our opinion most appropriate for the model Hamiltonian (1) where the \( f \)-states are taken to be non-degenerate (s-type). Without using the full orbital degeneracy, one could alternatively use the g-factors of the real materials. Other authors have even completely neglected the coupling of the magnetic field to conduction band states arguing that the corresponding g factor is negligible.

We use the dynamical mean-field theory (DMFT) in combination with the modified perturbation theory (MPT) to determine the one-electron Green’s function, from which the excitation spectrum as well as magnetization, effective mass and other quantities can be calculated. This method has previously been applied to the paramagnetic PAM and the ferromagnetic PAM, so we can confine ourselves to a short summary: The underlying idea of the DMFT is that the local self-energy such as occurs in the limit of infinite spatial dimension, \( \Sigma_{\text{loc}} \), can be taken to be that of an appropriately defined single-impurity Anderson model.

We solve the latter using the modified perturbation theory. Our starting point is the following ansatz for the on-site Coulomb interaction strength between two \( f \)-electrons. Throughout this paper, the conduction band will be described by a free (Bloch) density of states, \( \rho_0(E) = \frac{1}{N} \sum_{\epsilon} \delta(E - \epsilon(\vec{k})) \), of semi-elliptic shape. Its width \( W = 1 \) sets the energy scale, and its center of gravity the energy-zero: \( T_{ii} = \frac{1}{N} \sum_{\epsilon} \epsilon(\vec{k}) = 0 \). The magnetic field \( b_{\text{ext}} \) is given in energy units of the band width \( W \) (\( z_\sigma = +1(-1) \) for \( \sigma = \uparrow(\downarrow) \)). The external field couples equally to the spin of the \( f \) and conduction band electrons. This is in our opinion most appropriate for the model Hamiltonian (1) where the \( f \)-states are taken to be non-degenerate (s-type). Without using the full orbital degeneracy, one could alternatively use the g-factors of the real materials. Other authors have even completely neglected the coupling of the magnetic field to conduction band states arguing that the corresponding g factor is negligible.

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Using the perturbation theory around the Hartree-Fock solution introduces an ambiguity into the calculation. Within the self-consistent Hartree-Fock calculation, one can either choose the chemical potential to be equivalent to the chemical potential of the full MPT calculation, or take it as parameter \( \tilde{\mu} \) to be fitted to another physically motivated constraint. In reference [17] the Luttinger theorem, or equivalently the Friedel sum rule [18], was used to determine \( \tilde{\mu} \). As discussed in Ref. [13], we use the physically motivated condition of identical impurity occupation numbers for the Hartree-Fock and the full calculation \( \langle n_i^{\text{HF}} \rangle = \langle n_i^{(f)} \rangle \) to determine \( \tilde{\mu} \), which also allows for a consistent extension of the method to finite temperatures [11, 22]. The result not only describes in detail in references [11, 22], we make use of the moments of the spectral density. This procedure is described in detail in references [11, 22]. The result not only fulfills the \( V = 0 \) limit, but also recovers the high-energy behavior of the Green’s function up to the order \( \frac{1}{\epsilon} \).

It has been shown that the approximation scheme, as described above, gives qualitatively reliable results by comparing with numerical renormalization group theory and quantum Monte Carlo calculations for two different “strong-coupling effects”, namely the Mott-Hubbard insulator and ferromagnetism in the single-band Hubbard model. The periodic Anderson model was investigated by this and related methods in the paramagnetic and the ferromagnetic phase [13].

In the following we present results for the periodic Anderson model in the paramagnetic, but close to the ferromagnetic region of the phase diagram [13].

In Fig. 1, the f-magnetization \( m^{(f)} = \frac{n^{(f)}_\uparrow - n^{(f)}_\downarrow}{n^{(f)}_\uparrow + n^{(f)}_\downarrow} \) and the (spin-dependent) quasiparticle weight \( Z_\sigma = (m^{*})^{-1} = (1 - \frac{\partial^2 E}{\partial \mu^2} |_{\mu = 0})^{-1} \) are plotted as function of the external field \( b_{\text{ext}} \) for \( \epsilon_f = -0.4, U = 4, n^{(\text{tot})} = 1.85, V = 0.2 \) and zero temperature. These parameters away from the symmetric point (where \( \epsilon_f = -\frac{U}{2} \)) are close to the ferromagnetic phase which would be reached for lower electron density \( n^{(\text{tot})} \lesssim 1.7 \) [13]. The increase of \( m(b_{\text{ext}}) \) (thick solid line) shows a sharp rise at \( b_{\text{ext}} = b^{*}_{\text{ext}} \approx 0.033 \). We call this phenomenon a metamagnetic transition. In Fig. 2, a discontinuous jump in \( n^{(f)} \) is visible. However, due to the finite numerical resolution of our calculations we cannot rule out a continuous transition. For a range of \( b_{\text{ext}} < b^{*}_{\text{ext}} \) a high-magnetization and a low-magnetization self-consistent solution co-exist. For \( T = 0 \), the latter is always stable. The spin-up quasiparticle weight vanishes at \( b^{*}_{\text{ext}} \) implying a maximum in the effective mass. For \( b_{\text{ext}} > b^{*}_{\text{ext}} \), the quasiparticle weight is significantly larger than for \( b_{\text{ext}} = 0 \). As shown in the inset, the metamagnetic transition is also accompanied by a sharp increase in \( Z_\sigma \) at \( b_{\text{ext}} = b^{*}_{\text{ext}} \).

In Fig. 2 the magnetization and the quasiparticle weights are shown as function of interaction strength \( U \) for fixed \( b_{\text{ext}} = 0.1 \). At \( U = U^{*} \) a metamagnetic transition is also observable, being accompanied by the same behavior of \( Z_\sigma \) as discussed above.

The \( j \)-DOS for \( b_{\text{ext}} \in \{0.0, 0.02, 0.04, 0.06\} \) is plotted in Fig. 3 for the same parameters used in Fig. 1. For zero-field, the DOS consists of lower charge excitation at \( \epsilon_f \), upper charge excitation at \( \epsilon_f + U \) and the Kondo resonance at \( E \approx \mu \) which is split by the coherence gap. Applying a small external magnetic field \( (b_{\text{ext}} \leq 0.04) \) induces a Zeeman shift proportional to \( b_{\text{ext}} \), which is best visible in the upper charge excitation. The Kondo resonances in the spin-up and down channel seem also to be shifted, however, in the opposite direction: Spin-up to higher, and spin-down DOS to lower energies (“inverse Zeeman shift”). This is very unexpected behavior and is only found for parameters that show a metamagnetic transition. For other parameters further away from the ferromagnetic phase (e.g. \( U < U^{*} \) or also at the symmetric point \( \epsilon_f = -\frac{U}{2} \)) a “normal” Zeeman shift is observed for the Kondo resonance. Closer investigation shows that the apparent shift is indeed no shift, but a suppression of spectral weight in the spin-up channel just below the Fermi energy. This suppression is also indicated by
the paramagnetic DOS where the DOS for lower fields more or less correspond DOS strongly resemble those of the ferromagnetic PAM 13 chemical potential is visible in the spin-↓ spin-channels only. These DOS strongly resemble those of the ferromagnetic PAM whereas the DOS for lower fields more or less correspond to the paramagnetic DOS plus the Zeeman shift. So the metamagnetic transition closely resembles the para-to ferromagnetic transition. This agrees with the fact that the metamagnetic transition is clearly a strong coupling phenomenon as was also found for ferromagnetism in the intermediate-valence regime (see Fig. 3).

Let us shortly discuss the nature of the metamagnetic transition exhibited by our results. The origin of the transition is somewhat different from previously discussed approaches to metamagnetism, such as the Kondo-volume collapse approach, or explanations based on special features of the density of states. For example, solving the PAM within self-consistent perturbation theory, one also finds a metamagnetic transition. Here, however, the transition originates from sharp features in the excitation spectrum which are already present for zero-field. On applying the field, the Zeeman shift can push these features across the chemical potential and thus dramatically change the spin-dependent f-occupation and hence the magnetization. In our results however, the transition can be traced back to dramatic changes of the correlation-induced features of the excitation spectrum due to the external magnetic field. This clearly distinguishes the scenario presented in this paper from previously discussed metamagnetic transitions in the PAM. This manifests itself furthermore in the observed hysteresis which cannot occur in a scenario where the transition is due to the Zeeman shift of a strongly peaked DOS.

A phenomenologically similar metamagnetic transition is known to exist for the half-filled Hubbard model. Here, a jump in the magnetization as function of external field is found for interaction strengths U close to the critical Uc separating the Mott-insulator from the metallic regime. Similar to the transition described in this paper, the transition in the Hubbard model also shows a hysteresis and is indicated by a suppression of the Kondo resonance. Another similarity might be seen in the fact that in both metamagnetic transitions, the high-magnetization state is characterized by a stronger localization of the correlated electrons. In the Hubbard model, this transition is found for half-filling where it leads from a metallic to an insulating state. In the transition presented here we also see a tendency towards stronger localization of the f-electrons. Contrary to the situation in the half-filled Hubbard model, however, the PAM is metallic above and below the critical field. Furthermore, the suppression of the Kondo resonance occurs only in the proximity of the ferromagnetic phase, and only in the spin-↑ channel.

The metamagnetic transition is not simply an enlargement of the 'ferromagnetic' phase caused by the magnetic field. If this were the case one would expect similar behavior using other approximation methods, which yield almost the same ferromagnetic phase diagram as the MPT such as the modified alloy analogy and the spectral density approximation for the PAM. However, both methods do not show the metamagnetic transition. The low-energy (‘Kondo’) physics, which are not recovered by the other methods play a decisive role.

What is the relevance of our results to the metamagnetic transitions observed experimentally? The most well-known example of a heavy-fermion compound showing a metamagnetic transition from para- to a “ferromagnetic” state is CeRu2Si2. This material exhibits peculiar behavior alongside the metamagnetic transition. The effective mass shows a sharp maximum at \( b_{\text{ext}} = b^*_{\text{ext}} \), and for \( b_{\text{ext}} > b^*_{\text{ext}} \) is suppressed, which is exactly the behavior of the metamagnetic transition of the PAM discussed above. Furthermore, the experimentally confirmed stronger localization of the f-electrons in the high-magnetization state is indicated by the jump in \( n(\uparrow) \) that we discussed above.

There are two findings for CeRu2Si2, which are not reproduced by our approximation. Neutron scattering experiments revealed antiferromagnetic inter-site correlations for \( b_{\text{ext}} < b^*_{\text{ext}} \). These correlations can not be found within our approximation scheme since it is based on dynamical mean-field theory. However, the parameters we have used above are close to the antiferromag-

FIG. 3: \( f\)-DOS for the parameters of Fig. 1 with \( b_{\text{ext}} \) as indicated in the different rows. The solid (dotted) line corresponds to the spin-\( \uparrow \) (\( \downarrow \)) channel. The two columns show the same DOS at different energy ranges: lower charge excitation and Kondo resonance in the left and the upper charge excitation in the right column. The chemical potential is given by the arrows.

a decrease of \( Z_\uparrow \) as seen in Fig. 1.

For fields above the metamagnetic transition, the picture changes dramatically. The lower charge excitation is fully saturated and the shift between the two spin channels of the upper charge excitation is much larger than one would expect from an extrapolation from the other figures. Finally, the Kondo resonance disappears in the spin-\( \uparrow \) DOS. A broad structure located well above the chemical potential is visible in the \( \downarrow \)-channel only. These DOS strongly resemble those of the ferromagnetic PAM whereas the DOS for lower fields more or less correspond to the paramagnetic DOS plus the Zeeman shift. So the metamagnetic transition closely resembles the para-to ferromagnetic transition. This agrees with the fact that the metamagnetic transition is clearly a strong coupling phenomenon as was also found for ferromagnetism in the intermediate-valence regime (see Fig. 3).

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netic regime of the PAM. It seems therefore reasonable to expect antiferromagnetic correlations in the zero-field (low-magnetization) state. The second effect is the volume effect of the metamagnetic transition. A Kondo-volume collapse similar to that discussed in connection with the $\gamma \to \alpha$ transition of CeRu$_2$Si$_2$ has been put forward as possible source of the metamagnetic transition in CeRu$_2$Si$_2$. Our model does not include any coupling to the lattice. However, the significant change in $f$-occupancy that we find should result in a volume change, if the lattice coupling were to be included. This could even amplify the transition.

Contrary to our results, the metamagnetic transition in CeRu$_2$Si$_2$ does not show hysteresis. Let us point out that in our results we find merely a “mathematical hysteresis”. In Fig. 3 the thick line is the physically meaningful result whereas the thin line represents only a mathematical solution to the equations to which we cannot ascribe a clear physical meaning. However, one could assume that for finite temperatures, a true hysteresis would be found. Experimentally, a metamagnetic transition from a paramagnetic state that is accompanied by a hysteresis is realized in UCOT. This material, however, does not show a sharp maximum in the effective mass of the quasiparticles, so it is unclear whether the picture described in this paper is relevant for this material.

At this point, let us comment on the critical fields necessary to drive the metamagnetic transition. In our units, $b_{\text{ext}} = 0.033$ corresponds to several hundreds of Tesla if the bandwidth would be of order 1 eV. This is much too large compared to the above-cited experimental results. We believe this is a limitation of the MPT approximation. In our explanation, the transition is closely connected with the low-energy properties, the existence and the width of the Kondo resonance. The MPT, however, tends to overestimate the low-energy scales, so the absolute values of the critical field as obtained by our calculation should not be taken too seriously.

To summarize, we have presented a new scenario for a metamagnetic transition based on a strong electron-electron coupling effect. For a periodic Anderson model, with parameters located in the paramagnetic regime between the ferro- and the antiferromagnetic phase, an increase of the external field for sufficiently high interaction strength leads to a sudden sharp increase of the magnetization (metamagnetic transition). This is accompanied by a suppression of low-energy spectral weight (Kondo resonance) in the spin-$\uparrow$ DOS. The features of the transition show strong similarities to the experimentally observed metamagnetic transition of CeRu$_2$Si$_2$. The decreasing spectral weight near the Fermi level prior to the metamagnetic transition could serve as an experimentally accessible indicator for the relevance of the proposed mechanism to the real physics of CeRu$_2$Si$_2$.

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