Field-induced suppression of the heavy-fermion state in YbRh$_2$Si$_2$

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Abstract. We present calculations of the magnetic-field-induced changes of the heavy quasiparticles in YbRh$_2$Si$_2$ which are reflected in thermodynamic and transport properties. The quasiparticles are determined by means of the Renormalized Band Method. The progressive de-renormalization of the quasiparticles in the magnetic field is accounted for using field-dependent quasiparticle parameters deduced from Numerical Renormalization Group studies. Consequences for the interpretation of experimental data are discussed.

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1. Introduction

Magnetic fields may strongly affect the electronic properties of materials containing lanthanide or actinide ions [1]. In these systems, the relevant energy scales of the electronic system are strongly reduced due to the strong correlations of the partially filled 4f shells (for recent reviews see [2, 3] and references therein). The small energy scales comparable to a Zeeman energy of \( \sim 6 \) meV at 50T arise from removing local degeneracies by the anisotropy of the crystalline electric field (CEF), from building-up long-range order among moments or from forming (local) singlets via the Kondo effect. Important examples for unusual behavior are the field-induced changes of carrier-concentration in CeB\(_6\) and CeBiPt [4, 5, 6, 7] and their effective masses in filled Pr skutterudite compounds [8, 9] which result from the interplay of the magnetic and the low-energy CEF states. In the heavy fermion system (HFS) CeRu\(_2\)Si\(_2\), a deHaas-vanAlphen (dHvA) frequency changes abruptly at the metamagnetic transition [10]. This reflects changes in the ground state from a Fermi liquid with f-derived itinerant quasiparticles and a “large” Fermi surface to a conventional metal with polarized local f-moments and, concomitantly, a “small” Fermi surface [11, 12, 13, 14]. Metamagnetic transitions in Ce-based heavy-fermion compounds are a topic of high current interest [15].

The temperature vs. magnetic-field phase diagram of YbRh\(_2\)Si\(_2\) exhibits numerous anomalies [16]. This heavy fermion compound which crystallizes in the tetragonal ThCr\(_2\)Si\(_2\) structure has been in the focus of interest during the past decade because it has emerged as a prototypical system for investigation of quantum critical phenomena [17]. In its ground state, YbRh\(_2\)Si\(_2\) orders antiferromagnetically below the Nel temperature, \( T_N=70 \) mK [18]. By applying a weak magnetic field of \( B_c=60 \) mT in the basal plane the magnetic order is suppressed [19] and the characteristic features of a Fermi liquid, e. e., a specific heat varying linearly with temperature, a temperature-independent magnetic susceptibility with the Sommerfeld-Wilson ratio of order unity and an electrical resistivity \( \Delta \rho(T) = \rho(T) - \rho(0) \sim T^2 \) are observed. The quasiparticles are derived from the 4f degrees of freedom which have to be included in the “large” Fermi surface.

The present paper focuses on the magnetic-field induced variations and anomalies in the Fermi liquid state [20]. At low temperatures the specific heat coefficient and the magnetic susceptibility both progressively decrease with magnetic field. Since the ratio of the two quantities remains constant we are lead to the assumption that the system remains in a Fermi liquid state. The suppression of the local Kondo effect and the resulting de-renormalization of the heavy quasiparticles, however, should lead to a smooth variation evolution with the field. It cannot explain the anomaly observed at \( \sim 10 \) T in various thermodynamic and transport properties [16]. On the other hand, a dramatic Fermi surface reconstruction as it would occur at a Lifshitz transition [21, 22] is not supported by the observed dHvA data [23, 24, 25].

We conjecture that the above-mentioned anomalies result from a combination of a coherence effect of the periodic Kondo lattice, i. e., a van-Hove-type peak in the
quasiparticle density of states (DOS) and a local many-body effect related to breaking-up the Kondo singlets. The qualitative picture is that the DOS splits in the presence of a magnetic field. The pronounced anomalies in the thermodynamic and transport properties are related to the fact that the van-Hove singularity moves through the Fermi energy. The changes are related to a redistribution of quasiparticles among the Zeeman-split sheets of the Fermi surface with extremely high effective masses which have not been observed in dHvA experiments so far. The topology of the observed sheets with relatively light quasiparticles is only weakly affected by the external field.

To confirm this hypothesis we investigate the evolution of the Fermi liquid state with an external magnetic field. The strongly renormalized quasiparticles cannot be calculated by standard band structure methods based on Density Functional Theory (DFT). We therefore adopt the Renormalized Band (RB) method which successfully describes the quasiparticle dispersion in Ce-based HFS [12, 26, 27, 28, 29, 30]. The procedure has to be slightly modified for Yb compounds which can be considered as the hole analogues of the Ce systems. The Fermi surfaces and effective masses deduced from RB calculation reproduce the Hall effect data of the isostructural and isoelectronic HFS counterpart YbIr$_2$Si$_2$[31].

The paper is organized as follows: In Section II we shall delineate the RB method. The details of the calculational scheme are given in Section III. We discuss the results in Section IV and close with a summary and an outlook in Section V.

2. Renormalized Band method

The strongly renormalized heavy quasiparticle bands are determined by means of the Renormalized Band scheme which combines material-specific ab-initio methods and phenomenological considerations in the spirit of Landau. A detailed description of the method is given in Refs. [11, 12]. The key idea is to construct an effective Hamiltonian for the low-energy excitations which uses the ab-initio potentials for the weakly correlated conduction electron channels while introducing a small set of parameters to account for the specific local correlations among the 4f-electrons. The calculation of the selfconsistent ab-initio potentials and the weakly correlated conduction bands will be described in the subsequent section. The renormalized parameters are determined once by fitting to experiment and are kept fixed during subsequent investigations. Operationally, the renormalization procedure amounts to transforming the f-states of the spin-orbit ground state multiplet at the lanthanide site into the basis of CEF eigenstates |m⟩ and introducing resonance-type phase shifts

\[ \hat{\eta}_{fm} (E) \simeq \arctan \frac{\hat{\Delta}_f}{E - \hat{\epsilon}_{fm}} \]

where the resonance width \( \hat{\Delta}_f \) accounts for the renormalized quasiparticle mass. The resonance energies \( \hat{\epsilon}_{fm} = \hat{\epsilon}_f + \delta_m \) refer to the centers of gravity of the f-derived quasiparticle bands. Here \( \hat{\epsilon}_f \) denotes the position of the band center corresponding
Figure 1. Renormalized Band method: Schematic comparison of typical quasiparticle DOS of Yb (upper panel) and Ce (lower panel) HFS. Since the number of 4f holes (Yb case) and electrons (Ce case) is slightly smaller than unity the corresponding band centers are below and above the Fermi energy, respectively.

to the CEF ground state while the $\delta_m$ are the measured CEF excitation energies. One of the remaining two parameters, $\tilde{\epsilon}_f$, is determined by imposing the condition that the charge distribution is not altered significantly by introducing the renormalization. This makes the RB method a single-parameter scheme. The free parameter, $\tilde{\Delta}_f$, is adjusted so as to reproduce the coefficient of the linear specific heat at low temperatures. The method has been shown to reproduce Fermi surfaces and anisotropies in the effective masses of a great variety of Ce-based compounds. In addition, it allows one to predict Fermi liquid instabilities. Typical results for Ce systems can be found in [2, 28, 32] and reference therein.

In calculating the coherent 4f-derived quasiparticle bands in the Yb-based heavy fermion compounds we essentially follow the procedure for the Ce-case as described above. We have to account for the fact that Yb can be considered as the hole analogue of Ce. Operationally this implies that we have to renormalize the 4f $j=7/2$ channels at the Yb sites instead of the 4f $j=5/2$ states in the Ce case. As the 4f hole count is slightly less than unity the center of gravity $\tilde{\epsilon}_f$ will lie below the Fermi energy. In addition, we have to reverse the hierarchy of the CEF scheme, i. e.,

$$\tilde{\epsilon}_f < 0 \quad ; \quad \tilde{\epsilon}_f m = \tilde{\epsilon}_f - \delta_m \quad .$$

These modifications are schematically summarized in Figure 1.
We should like to mention that the effective band structure Hamiltonian constructed along these lines corresponds to a hybridization model which closely parallels the one obtained from the periodic Anderson model in mean-field approximation.

In the presence of a magnetic field, the quasiparticles have to be described by field-dependent parameters for the level $\tilde{\epsilon}_{fm}(h)$ and the resonance width $\tilde{\Delta}_f(h)$. In the systems under consideration, the energy difference between the CEF ground state doublet and the excited states largely exceeds the resonance width which is of the order of the characteristic energy $k_B T^*$. The low-temperature properties derived from the heavy quasiparticles will reflect the spatial symmetry of the CEF ground state wave function. The energy $h = \frac{1}{2} g_{eff} \mu_B H$ denotes the (anisotropic) Zeeman splitting of the free ion CEF ground state which is related to that of an effective spin-1/2-system by introducing anisotropic effective $g$-factors. In general, we anticipate the variation with magnetic field of the parameters $\tilde{\epsilon}_{fm}(h)$ and $\tilde{\Delta}_f(h)$ to be highly non-trivial due to the strong correlations.

The variation with magnetic field of the phase shifts in the low-field limit was discussed by Nozieres [33] for dilute magnetic alloys. In his seminal paper he emphasized the importance of the molecular field correction. The latter accounts for the quasiparticle interactions which are reflected in the value $R = 2$ of the Sommerfeld-Wilson ratio. In our parametrization the molecular field correction translates into a correlation-enhanced Zeeman splitting of the centers of gravity $\tilde{\epsilon}_{fm}(h)$. The field-dependence of the renormalized band width $\tilde{\Delta}_f(h)$, however, cannot be derived from phenomenological considerations assuming “rigid” quasiparticles. In the present calculation, we use field-dependent parameters $\tilde{\epsilon}_{f}(h)$ and $\tilde{\Delta}_f(h)$ which are obtained from fits to field-dependent quasiparticle DOS of the single-impurity Anderson model [34, 35, 36, 37]. The latter are calculated microscopically by means of the Numerical Renormalization Group (NRG). This procedure properly accounts for the progressive de-renormalization of the quasiparticles with increasing magnetic field and the correlation-enhanced Zeeman splitting. As will be shown below, it allows for a quantitative description of the observed field-induced anomalies as well as for the observed dHvA data in the high-field regime.

3. Computational method

The band-structures were obtained by the fully relativistic formulation of the linear muffin-tin orbitals (LMTO) method [38, 39, 40]. The spin-orbit interaction is fully taken into account by solving the Dirac equation. We adopt the atomic sphere approximation (ASA) and include the combined correction term which contains the leading corrections to the ASA[38]. The calculations are done at the experimental lattice parameters $a=b=4.007\,\text{Å}, c=9.858\,\text{Å}$ for YbRh$_2$Si$_2$. We include s-p-d-f-components at the Yb and the transition metal (Rh, Ir) sites and s-p-d-components at the Si sites. We start by optimizing the density distribution of the conduction electrons for the magnetic configuration by treating 13 4f electron as part of the ion core. The fully relativistic
potential parameters for the non-states are determined in selfconsistent Dirac-relativistic
calculations. Although the relativistic effects hardly change the electron density
distribution they nevertheless influence the actual placements of the energy bands.
This aspect is particularly important for the renormalized band structure since the
soin-orbit splitting s of the d-states are rather large on the energy scales relevant for
the strongly renormalized heavy quasiparticles. Exchange and correlation effects were
introduced using the Barth-Hedin potential [41]. The band-structure was converged
for 405 Κ-points in the irreducible wedge, whose volume equals 1/16 of the Brillouin
zone. The density of states (DOS) was evaluated by the tetrahedron method with linear
interpolation for the energies. For the conduction band the DOS was calculated at 0.25
mRy (≈ 0.0034 eV) intervals.

The dispersion of the weakly-correlated non-f states is determined by a standard
band structure calculation assuming the the Yb ion is in 4f\textsuperscript{13} configuration. The effective
potentials are generated selfconsistently within the Local Density Approximation (LDA)
to Density Functional Theory (DFT). The strong Coulomb repulsion among the 4f
electrons which suppresses charge fluctuations is implicitly accounted for by treating
the 4f electrons as part of the ion core assuming that they do not hybridize with the
conduction states. We refer to this calculation as f-core calculation. The electronic
density of the 4f states is assumed to be that encountered in isolated atoms. This
assumption seems justified for the systems under consideration whose 4f valence deviates
only weakly from the integer value. Details and results are given in [31].

Before we turn to a discussion of the RB calculation let me mention a few
caveats concerning ab-initio LDA calculation: In Yb-based heavy-fermion compounds
we encounter new difficulties in addition to the well-known inadequacies observed in
Ce-HFS. The experimental 4f valence in the Yb-HFS under consideration is close to
13. Calculations show that there exists only a rather narrow pressure range in which
the 4f\textsuperscript{13} configuration is stable over the 4f\textsuperscript{14} configuration[42]. For the experimental
lattice constants the 4f\textsuperscript{14} configuration is the stable one. Ab-initio LDA calculation can
therefore tend to converge to a ground state where the 4f shell is filled and sitting below
the Fermi level[43]. Since the character of the states at the Fermi energy is not correctly
described we anticipate significant deviations in the effective masses in this case.

The calculations reported here adopt a CEF scheme which is consistent with
susceptibility and the inelastic neutron data [44, 45]. The latter indicate that the
4f\textsuperscript{13} states in YbRh\textsubscript{2}Si\textsubscript{2} are split into 4 doublets with the energies 0-17-25-43 meV.
The parameters for the tetragonal CEF are B\textsubscript{20}=0.5246 meV, B\textsubscript{40}=0.01195 meV,
B\textsubscript{60}=-0.0004725 meV, B\textsubscript{44}=0.03598 meV, B\textsubscript{64}=-0.01206 meV [46, 47]. The low-energy
properties are mainly determined by the CEF ground state which is a superposition
of |j = 7/2; j\textsubscript{z} = ±5/2) and |j = 7/2; j\textsubscript{z} = ±3/2) and which is well separated from the
excited states. The density distribution (hole representation) in Figure 2 suggests that
the coupling to the conduction states is rather weak. The resulting g-factors are \(g|| = 0.26\)
and \(g\perp = 3.79\) for magnetic fields parallel to the tetragonal axis and in the basal plane.

We use a quasiparticle resonance width of \(\tilde{\Delta}_f = 20K\) as inferred from specific heat
and thermopower measurements [48, 16].

4. Results

The RB calculation yields narrow bands with f-character in the vicinity of the Fermi energy while the dispersion of the broad non-f conduction bands remains essentially the same as in the local moment regime. The quasiparticle DOS displayed in Figure 3 is mainly due to the CEF-split 4f-states. The RB calculations yield a DOS of $\sim 290$ states/(eV unit cell) corresponding to specific heat coefficient $\sim 680 \text{mJ/(mole K}^2\text{)}$. This value should be considered as theoretical zero-field limit of the specific heat coefficient measured in the Fermi liquid state at small finite magnetic fields. The actual zero-field specific heat data are strongly enhanced by the anomalous fluctuations associated with the quantum critical point.

There are three bands intersecting the Fermi energy. In the following discussion, we shall neglect the small $\Gamma$-centered electron pocket and focus on the two bands giving rise to the two major sheets displayed in Figure 4. The overall topology qualitatively agrees with LDA results of Refs. [49, 50, 23, 24, 25]. The dominant contribution to the quasiparticle DOS and, concomitantly, to the specific heat and magnetic susceptibility comes from the Z-centered hole surface which has predominantly f-character. The states forming the “jungle gym”, on the other hand, are strongly hybridized. The anisotropy of the CEF ground state results in a strongly anisotropic hybridization. In the present context it is important to note that the dispersion of the heavy band is rather flat over a wide range of the Brillouin zone. As a result we find a very sharp van-Hove type feature in the quasiparticle DOS displayed in Figure 3. It is this feature which leads to the anomalies at $\sim 10T$.

We next turn to the variation with magnetic field of the quasiparticle states. The results for the DOS at the Fermi energy are summarized in Figure 5. They should be compared to the specific heat and susceptibility data [20]. It is instructive to separately estimate the relative importance of the pure band structure effects and the consequences of the local correlations. For this reason, we first calculate the energy bands and DOS assuming that the CEF ground state is rigidly split by the external field. To calculate the bare Zeeman splitting we use the effective $g$-factor for magnetic fields in the basal plane. The resulting DOS at the Fermi energy $D(0)$ is almost constant in the field of interest which is clearly at variance with the experimental findings. Including quasiparticle interactions via the molecular field correction reproduces a discontinuity at $H\sim 10T$. On the low-field side, however, this ansatz predicts an increase in $D(0)$ which cannot be reconciled with the observed behavior. The progressive de-renormalization of the quasiparticle mass, on the other hand, does explain the smooth decrease in the linear specific heat coefficient and the magnetic susceptibility, the sharp drop at $\sim 10T$, however, is absent. Only the full variation with field of the quasiparticle parameters gives a satisfactory description of the experimental data.

Finally we would like to comment on the dHvA results in high magnetic fields.
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Figure 2. YbRh$_2$Si$_2$: Contour of equal 4f hole density for the CEF ground state of Yb$^{+3}$. The spheres denote the nearest neighbor atoms of the central Yb in the tetragonal body centered structure.

Figure 3. YbRh$_2$Si$_2$: Zero-field quasiparticle DOS from RB calculation. The f-bands are clearly split by the CEF (left panel). The low-energy properties are determined by the peak at the Fermi energy which exhibits a “coherence gap” and a van-Hove type singularity (right panel).

Several groups reported a large extremal orbit corresponding to relatively light quasiparticles. The jungle gym sheet of the RB Fermi surface in Figure 6 is consistent with these findings. In a field of $\sim$15T there is a closed orbit in the 110 plane with F=13kT and $m^{*} \sim$20m which agrees well with the values given by Westerkamp[51]. From Figure 6 it is obvious that the closed orbit is confined to a relatively narrow range of magnetic field orientations. In addition, the orbit cannot be assigned to a “small” Fermi surface expected in the local moment regime. The calculated area of the large orbit is consistent with the quantum oscillation frequency found recently by Sutton et
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Figure 4. YbRh$_2$Si$_2$: Major sheets of the quasiparticle Fermi surface (zero-field RB calculation). It consists of a Z-centered hole surface (left panel) and a multiply-connected surface (“jungle gym”) (right panel).

Figure 5. YbRh$_2$Si$_2$: Variation with magnetic field in the basal plane of the quasiparticle DOS at the Fermi level D(0) derived from the RB calculation (filled circles). The field-dependent parameters $\tilde{\epsilon}_f(h)$ and $\tilde{\Delta}_f(h)$ are determined from fits to NRG calculations. Included for comparison are predictions assuming single-particle Zeeman splitting (dotted line), decreasing effective mass plus single-particle Zeeman splitting (dot-dashed line), correlation enhanced Zeeman splitting with zero-field effective mass (open diamonds).
al. [25] for field directions around the (110) direction. Since the topologies of their J-sheet and that of the RB calculation are rather similar I guess that then discrepancy in field orientation may be due to a different labelling of the axes. In the present paper we refer to the basis of the reciprocal lattice which is rotated by 45° relative to the one of the underlying crystal lattice.

5. Summary and outlook

We calculated the variation with magnetic field of the quasiparticle bands in YbRh$_2$Si$_2$ by means of the RB method. The field-dependent quasiparticle parameters extracted from a NRG treatment of the single-impurity Anderson model account for the correlation enhanced Zeeman splitting and the reduced effective mass of the CEF ground state doublet. The anomalies observed at ~10T in numerous thermodynamic and transport properties results from a van Hove-type structure which appears below the Fermi energy in the zero-field limit. The field-dependent f-derived quasiparticles and their “large” Fermi surface reproduce the variation with magnetic field of the linear specific heat coefficient and the magnetic susceptibility. The “large” Fermi surface consistently explains the quantum oscillation frequency observed in the high-field limit. In conclusion, the following picture emerges: To quantitatively understand the unusual behavior of YbRh$_2$Si$_2$ in the magnetic field we have to account both for local many-body effects and coherence structures from the periodicity of the lattice.
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References

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