High-Field ESR study of the Kondo lattice system YbRh$_2$Si$_2$

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Abstract. A multi-frequency electron spin resonance (ESR) study of the heavy fermion compound YbRh$_2$Si$_2$ has been performed in fields up to $\sim$ 8 T. In this regime the system is close to the breakdown of the heavy-fermion behavior which is confined to temperatures and fields $T < T_0 \approx 25$ K and $B < B^* \approx 10$ T, respectively. A well-defined strongly anisotropic ESR mode is observed. We find striking similarities between the properties of this signal and the behavior of the electronic specific heat and the $^{29}$Si NMR data which reflect the crossover between different electronic regimes in the Kondo state. These similarities suggest that the observed ESR mode corresponds to a "heavy electron spin resonance", i.e., a joint resonance excitation of the strongly hybridized $f$- and conduction electron states.

1. Introduction

In rare-earth (RE) Kondo lattice systems ($\text{RE} = \text{Ce, Yb}$) strong electron-electron correlations determine the low temperature physical properties. They are controlled by two competing interactions: the Kondo effect which yields the screening of local 4$f$-magnetic moments by conduction electrons (CE) and results in a nonmagnetic ground state and the so-called RKKY-interaction between the 4$f$-states and CE that stabilizes a magnetically ordered ground state. A prominent example of such class of systems is the intermetallic compound YbRh$_2$Si$_2$. Here the competition between these two interactions that can be tuned by a magnetic field $B$ and temperature $T$ yields different phases and crossover behaviors such as antiferromagnetic order, quantum criticality, heavy fermion- and non-Fermi liquid (NFL) behavior [1, 2, 3, 4, 5]. One of the surprising results on this material was the observation of a sharp anisotropic electron spin resonance (ESR) signal resembling ESR of localized Yb$^{3+}$ states [6]. However, the occurrence of this signal down to the lowest accessible temperature of 0.69 K [7] where the single ion Kondo effect is expected to screen the magnetic moments has suggested the non-local origin of the resonance [8]. One can conjecture that in YbRh$_2$Si$_2$ this signal is given by a joint resonance excitation of strongly hybridized 4$f$- and CE states, i.e. by a "heavy electron spin resonance". To verify this idea we performed high field ESR (HF-ESR) measurements close to the NFL/LFL-crossover and the breakdown of the heavy-fermion behavior which is confined to temperatures and fields $T < T_0 \approx 25$ K and $B < B^* \approx 10$ T, respectively [4] (for details, see Ref. [9]).
2. Experimental Setup

HF-ESR measurements were carried out on a high quality single crystal of YbRh$_2$Si$_2$ in a reflection geometry with a Millimeterwave Vector Network Analyzer from AB Millimetre at $\nu = 93 - 360$ GHz in a field range $B = 0 - 14$ T. Our HF-ESR setup allows to measure both the amplitude and the phase shift of the reflected signal (Fig. 1). This enables a separation of the absorption and dispersion part of the complex resonance response of a metallic sample. Simultaneous fitting of the amplitude and the phase shift at the resonance provides an accurate determination of the resonance field $B_{\text{res}}$ and the width $\Delta B$ of the resonance modes.

3. Results

In agreement with previous low-frequency ($\nu < 34$ GHz) low field ($B < 1$ T) ESR (LF-ESR) studies [6, 7] the HF-ESR signal can be observed for the $B \perp c$ geometry and is absent for the $B \parallel c$ orientation. For $B \perp c$ a complex ESR response can be fitted by single Lorentzian absorption and dispersion functions, both containing the same parameters $B_{\text{res}}$ and $\Delta B$ (Fig. 1). In Figs. 2 and 3 we show exemplarily the $T$-dependences of the $g$-factor $g = h\nu/(\mu_B B_{\text{res}})$ and $\Delta B$ for two selected frequencies $\nu = 93$ GHz and 297 GHz ($h$ and $\mu_B$ are the Planck constant and Bohr magneton, respectively). The corresponding resonance fields amount to $B_{\text{res}} \approx 1.8$ T and 6.15 T, respectively. At a fixed $\nu$, the $g$-factor (Fig. 2) increases approximately as $\ln(T)$ at high temperatures for both frequencies. The rate of this increase is smaller for the higher frequency/magnetic field and the $g(T)$ dependence shows a saturation tendency below $\sim 4 - 5$ K (marked by arrow in Fig. 2). The $\Delta B(T)$-dependence (Fig. 3) can be described as a sum of three contributions $\Delta B(T) = a + bT + c/(\exp(\Delta/T) - 1)$ similar to the LF-ESR analysis in Ref. [6]. Here $a$ depicts a $T$-independent contribution due to various kinds of inhomogeneities, $bT$ stands for the relaxation broadening via electronic degrees of freedom, and the last term has been assigned to a relaxation channel via an excited doublet state of Yb$^{3+}$ at an energy $\Delta$ above the ground state. In the previous LF-ESR studies [6, 7] and also at $\nu = 93$ GHz and $B_{\text{res}} \approx 1.8$ T in the present work the data points closely follow this dependence in the entire temperature range of study. However, further increase of the frequency and magnetic field gives rise to a deviation from this dependence at $T \lesssim 7$ K, namely $\Delta B$ begins to decrease more rapidly (marked by arrow in Fig. 3). This feature is more clearly seen in the plot representation $\Delta B$ vs. $T^2$ (Fig. 3). Remarkably such a hump in the $\Delta B(T)$-dependence occurs closely to the $T$-region where the $g$-factor begins to saturate (Fig. 2). Similar features in the $g(T)$ and $\Delta B(T)$ dependences are present also at other frequencies in the range $93 - 360$ GHz (for details, see [9]).

Figure 1. (color online) (a) - absorptive (amplitude) and (b) - dispersive (phase shift) parts of the HF-ESR signal at 249 GHz for $B \perp c$ (noisy curves). Solid lines (red online) are fits to the Lorentzian absorption and dispersion function, respectively. Grey inset shows a polar plot of the resonance signal. In this representation a circle corresponds to the complex Lorentzian function. (See the text)
4. Discussion
To discuss the properties of HF-ESR in YbRh$_2$Si$_2$ it is instructive to plot the characteristic crossover temperatures in the behavior of $g$ and $\Delta B$ on the general phase diagram of this compound (Fig. 4). Of a particular interest is the crossover line that separates the electronic regime of the development of the heavy fermion state below the Kondo temperature $T_0 \approx 25$ K followed by the formation of the NFL behavior at higher $T$ from the LFL regime at lower $T$ and higher $B$ (broad line and full symbols in Fig. 4). The HF-ESR crossover temperatures (open symbols in Fig. 4) join this line and continue it towards the crossover line for the breakdown of the heavy fermion state (gray dash line in Fig. 4). In particular, the HF-ESR points overlap with the crossover points deduced from the measurements of the electronic specific heat $C_{el}$ which is a measure of the quasiparticle density of states $N(E_F)$ at the Fermi level $E_F$ [2]. The latter points mark the change of the $T$ behavior of the Sommerfeld coefficient $\gamma = C_{el}/T \propto N(E_F)$ from $-\ln T$ at higher $T$ to $\approx$ const below $T \sim 2 - 4$ K (Fig. 2, inset, and Ref. [2]). This has been interpreted as the development of heavy fermion quasiparticles at higher $T$ followed by the establishment of the LFL state at lower $T$. Moreover, one can trace similarities between the changes in the behavior of $\Delta B(T)$ at low $T$ and of the longitudinal $^{29}$Si spin relaxation rate $1/T_1$ divided by $T$, which is proportional to the momentum $q$ averaged dynamical electron spin susceptibility $\chi''(q, \omega)$ [3]. Such remarkable correspondences strongly suggest that HF-ESR in YbRh$_2$Si$_2$ is related to the resonance response of the heavy fermions in a magnetic field to the microwave radiation. One may suppose that a strong coupling between the conduction electrons and the local moments may yield a joint ESR response, plausibly of the same origin as recently observed in the ferromagnetic Kondo lattice system CeRuPO [10]. The heavy quasiparticles
inheret to a significant extent the anisotropic properties of the $f$-electrons, specifically a typical strong anisotropy of the $g$-factor. In this situation the uniform static ($\omega = 0$) spin susceptibility of heavy fermions contributes to the shift of the $g$-factor yielding a $T$-dependence similar to that of the electronic specific heat. In fact, the field dependence of the $g$-factor’s temperature variation, Fig. 2, reflects what qualitatively would be expected for $4f$ magnetic moments which are influenced by the Kondo effect: with increasing temperature and/or increasing magnetic field the Kondo interaction between $4f$ and conduction electron spins gets suppressed and the $g$-factor approaches values which correspond to the completely localized $4f$ spin.

5. Conclusions

In summary, we have studied HF-ESR in YbRh$_2$Si$_2$ in a $T - B$-parameter domain extending from the breakdown of the heavy fermion state at large $T$ and large magnetic fields until the crossover to the LFL state at lower temperatures. The features identified in the $T$-dependences of the $g$-factor and the ESR linewidth correspond closely to the crossover line that separates these different regimes. The sensitivity of HF-ESR to this electronic crossover suggests that the ESR response is given by a joint resonance excitation of the strongly hybridized $4f$- and itinerant electron states, i.e. to the “heavy electron spin resonance” and as such it directly probes the evolution of the Kondo state in this Kondo lattice system.

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6. References

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