Evolution of the Kondo State of YbRh$_2$Si$_2$ Probed by High-Field ESR

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An electron spin resonance (ESR) study of the heavy fermion compound YbRh$_2$Si$_2$ for fields up to
$\sim$8 T reveals a strongly anisotropic signal in the Kondo state below $\sim$25 K. A similarity between the T
dependence of the ESR parameters and that of the specific heat and the ^{29}Si nuclear magnetic resonance
data gives evidence that the ESR response is given by heavy fermions. Tuning the Kondo effect on the $4f$
states with magnetic fields $\sim$2–8 T and temperature 2–25 K yields a gradual change of the ESR g factor
and linewidth which reflects the evolution of the Kondo state in this Kondo lattice system.

Strong electron-electron (EE) interactions in metals yield a fascinating variety of novel and often interrelated quantum phenomena, such as quantum phase transitions, breakdown of the Landau Fermi liquid (LFL) state, and unconventional superconductivity (for an overview see, e.g., [1]). In intermetallic compounds where $4f(5f)$ magnetic ions (e.g., Yb, Ce, U) build up a regular Kondo lattice, strong EE correlations are established by the coupling of local f magnetic moments with the conduction electrons (CE). As a consequence, a large effective mass enhancement of the quasiparticles (QPs) hallmarks the properties of paramagnetic heavy fermion metals. A competing interaction, the so-called RKKY interaction between the local f states via the sea of CE, favors a magnetically ordered ground state.

An important realization of a system where the delicate balance between Kondo and RKKY interactions can be investigated is the intermetallic compound YbRh$_2$Si$_2$, where antiferromagnetic (AF) order, quantum criticality, and heavy fermion and nonLFL (NFL) behavior can be tuned by a magnetic field B and temperature T [2–6] (Fig. 1). In the parameter domain where these remarkable electronic crossovers take place, a strong hybridization of $4f$ electrons with CE significantly broadens the otherwise atomically sharp f states. That is why the observation of a narrow electron spin resonance (ESR) signal in the Kondo state of YbRh$_2$Si$_2$ was very surprising [7]. While the reported pronounced anisotropy of the signal is indeed in accordance with an ESR of localized Yb^{3+} $4f$ moments, a nonlocal picture is suggested by the observation of this signal down to the lowest accessible temperatures of 0.69 K [8] where the single ion Kondo effect is expected to screen the magnetic moments. On the other hand the conduction electron ESR seems also unlikely because in this compound comprising heavy metal elements the spin-orbit (SO) coupling drastically shortens the electron spin lifetime [9].

To unravel a controversial nature of this resonance response we have studied ESR of a high quality single crystal of YbRh$_2$Si$_2$ in a broad magnetic field region varying between the low-field/low-frequency (LF) ($\sim$0.2 T/$\nu \sim$ 9 GHz) and the high-field/high-frequency (HF) ($\sim$8 T/$\nu \sim$ 360 GHz) regimes. In the LF limit YbRh$_2$Si$_2$ is in the NFL state, whereas in the HF regime it is in a LFL state close to the breakdown of the heavy fermion behavior
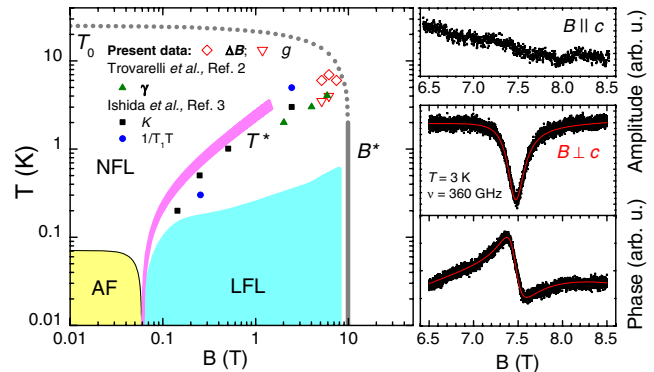

FIG. 1 (color online). Left: Schematic phase diagram of YbRh$_2$Si$_2$ for $B \perp c$ from Ref. [1]. Dashed and solid gray lines delineate the region of heavy QP behavior confined to the region $T < T_0$ and $B < B^*$ [2,4,5]. The LFL region denotes the T-B domain where the resistivity follows LFL behavior $\rho \propto T^2$. The broad (pink online) line T^* depicts the position of crossover in the isothermal Hall resistance, magnetostriction, magnetization, and longitudinal resistivity. Closed symbols depict $T(B)$-crossover temperatures below which specific heat (γ) [2] and ^{29}Si-NMR quantities [K and $1/(T_1 T)$] [3] become T independent. Open symbols display crossover temperatures from HF ESR $g(T, B)$ and $\Delta B(T, B)$ dependences (see text). Right: A representative HF ESR signal for the $B \perp c$ axis. Absorptive (amplitude) and dispersive (phase shift) parts of the ESR signal (noisy curves) are simultaneously fitted to a Lorentzian function (solid lines). No signal can be observed for the $B \parallel c$ axis, suggesting a strong g factor anisotropy.

which is confined to temperatures and fields $T < T_0 \approx$ 25 K and $B < B^* \approx 10$ T [4]. Here T_0 denotes the characteristic spin fluctuation temperature (which corresponds to the single ion Kondo temperature T_K) [2], and B^* is a characteristic field which separates more itinerant from more localized behavior of the $4f$ states [2,4,5]. In this parameter domain electronic specific heat (related to the density of states of QPs) [2,5] and nuclear magnetic resonance (NMR) data (related to the spin dynamics of QPs) [3] indicate the development of heavy fermion behavior with a field dependent crossover to clear LFL properties at low T [5]. We find that this development and crossover behavior is reflected also in the temperature and field dependence of the ESR response. This similarity gives evidence that ESR in YbRh$_2$Si$_2$ is given essentially by the resonance of heavy fermions, providing thus *direct* experimental access to the dynamics of heavy quasiparticles in the Kondo state.

LF ESR was measured with a standard highly sensitive cavity based reflection technique [7]. HF ESR was carried out in a reflection geometry with a millimeter wave vector network analyzer (AB Millimetre) at $\nu = 93$–360 GHz in a field range 0–14 T. We used a very sensitive induction mode scheme that detects the change of the microwave's polarization at the resonance [10].

The strong anisotropy of the ESR response corresponds to the strong magnetic anisotropy of YbRh$_2$Si$_2$ [2,11] and is nicely reflected in the HF ESR data: for the $B \parallel c$ axis no ESR response has been found, whereas a well defined resonance line with a g factor ~ 3.5 has been observed for $B \perp c$ (Fig. 1, right). Note that our HF ESR setup allows measurement of both the amplitude and the phase shift of the signal. 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 (Fig. 1, right) provides an accurate determination of the resonance field B_{res} and the linewidth ΔB.

The T dependences of the g factor $g = h\nu/\mu_B B_{res}$ and ΔB at selected excitation frequencies are summarized in Figs. 2 and 3. For large B_{res} the data appreciably scatter owing to a moderate signal-to-noise ratio, caused by a considerable broadening of the signal with increasing temperature and magnetic field strength, as well as the reduction of the microwave penetration depth with increasing ν. However, two central results can be deduced: (i) the g factor data sets show a decreasing T variation with increasing magnetic field, and (ii) one can identify distinct regimes with different T dependences of the g factor and ΔB in the data sets at $\nu = 249$, 297, and 360 GHz ($B_{res} \approx 5.15$, 6.15, and 7.45 T). At a fixed field (or frequency), the g factor [Fig. 2(a)] increases approximately as $\ln(T)$ at high T's for all fields. However, $g(T)$ shows a saturation tendency below ~ 4–5 K for the data at $\nu = 249$ and 297 GHz. At these frequencies the $\Delta B(T)$ data also show
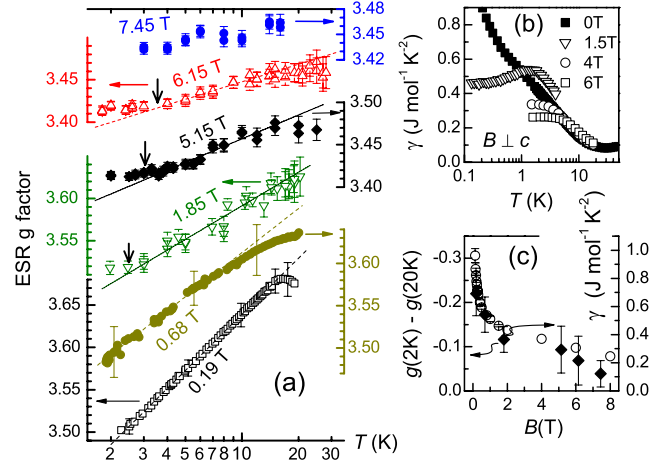

FIG. 2 (color online). (a) $g(T)$ dependence for $B \perp c$ at $\nu = 9$, 34, 93, 249, 297, and 360 GHz (from bottom to top), which correspond to resonance fields as indicated. Arrows indicate a crossover from a $-\ln T$ to $g = $ const behavior. (b) T dependence of the Sommerfeld coefficient γ at different magnetic fields (Ref. [5]). (c) Field dependence of low temperature γ (Ref. [5]) and g shift between 20 K and 2 K.

an anomaly, namely, a broad hump at somewhat higher $T \sim 7$–8 K [Fig. 3(a)].

As a function of temperature and magnetic field, the thermodynamic quantities, such as the electronic specific heat $C_{el} \propto N(E_F)T$ and the Pauli susceptibility $\chi_p \propto N(E_F)$, both being a measure of the density of states $N(E_F)$ of QPs at the Fermi level E_F and thus their effective mass m^*, experience notable modifications. In the T-B range of the HF ESR data χ_p probed by the ^{29}Si Knight
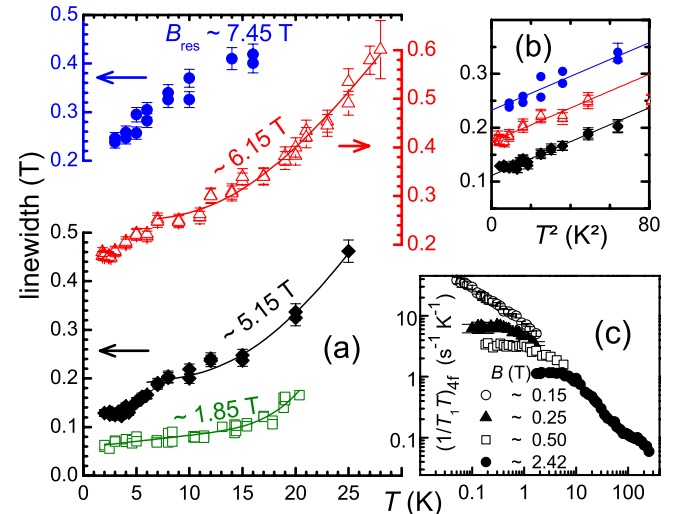

FIG. 3 (color online). (a) T dependence of the linewidth ΔB for $B \perp c$ at $\nu = 93$ (squares), 249 (diamonds), 297 (triangles), and 360 GHz (circles). Solid lines are fits (see the text). (b) Corresponding low-T data in the representation ΔB vs T^2. Solid lines are fits to the T^2 dependence. (c) T dependence of ^{29}Si relaxation rate $1/(T_1 T)$ from Ref. [3].

shift $K \propto \chi_p$ and the Sommerfeld coefficient $\gamma = C_{el}/T$ change their T dependences from $-\ln T$ at higher T to $\approx$ const below $T \sim 2$–4 K [Fig. 2(b) and Refs. [2,3,5]]. This may be interpreted as the development of the heavy fermion QPs at higher T followed by the establishment of the coherent LFL state at lower T. Remarkably, these changes occur at approximately the same temperatures and fields where the $\ln T$ dependence of the g factor levels off [$T \sim 2$–4 K; see arrows in Fig. 2(a) and symbols in Fig. 1, left]. A close correspondence between the increase of γ which reflects a strong enhancement of m^* and the g shift at low T can be seen in Fig. 2(c). Moreover, at low T there is an apparent correspondence between the changes of the T behavior of the dynamic quantities, namely, 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 $\chi''(q, \omega)$, and the ESR linewidth $\Delta B(T)$ (Fig. 3 and Ref. [3]). The crossover temperatures from specific heat, NMR, and present HF ESR data are summarized in the phase diagram in Fig. 1. The ESR points obviously fall into a common crossover line. Such remarkable similarities between the characteristic changes of γ and NMR quantities on the one hand and the HF ESR data on the other hand strongly suggest that ESR in YbRh$_2$Si$_2$ is given by the response of the heavy electrons to microwaves and as such it reflects a crossover between the different electronic regimes. Note that the diffusion time τ_D of the heavy electrons in YbRh$_2$Si$_2$ is long enough [12] for the ESR line shape to be Lorentzian, which is a limiting case of a "Dysonian" at $\tau_D \to \infty$.

In the following we discuss plausible reasons for making such a heavy electron spin resonance experimentally observable and its relation to the Kondo state of YbRh$_2$Si$_2$. Generally, ESR is given by the frequency dependent uniform ($q = 0$) transverse electron spin susceptibility. From its pole one obtains the resonance frequency $\nu = g\mu_B B_{res}/h$ and the linewidth $\Delta B \propto 1/T_2^*$. Here T_2^* is the electron spin-spin dephasing time, which in metals is equal to the spin-lattice relaxation time T_1 (see, e.g., Ref. [13]). For the spin resonance of conduction electrons (CESR) ν and ΔB depend on the details of the band structure and the SO interaction [9]. Owing to the large magnitude of the latter in metals containing heavy elements, the CESR signal becomes unobservable [9]. Magnetic resonance of the localized states in a metallic host can be observed much more easily even in materials with strong SO interaction. This is because the $4f$ electrons being buried deeply beneath the outer electron shells of the rare-earth ion retain the sharp atomiclike character of the energy states and generally mix little with the CE. Also in Kondo systems at $T > T_K$, where the local moments are still well defined, their resonance can be observed experimentally, as, e.g., in a dilute Au:Yb alloy with $T_K \sim 10$ μK [14]. However, at $T \lesssim T_K$ the signal is expected to disappear owing to the Kondo screening of the local moments by CE [7,12]. A single ion Kondo scenario for the occurrence of a sharp ESR mode of localized Yb^{3+} moments is unlikely for a regular Kondo lattice [15]. It is also unreasonable to discuss the ESR signal considering only conduction electrons, as if YbRh$_2$Si$_2$ were an uncorrelated metal. In this scenario a strong SO scattering due to the presence of heavy elements would smear out the resonance. In the Kondo state a coherent many-body interaction between itinerant electrons and local moments gives rise to QPs with a strongly enhanced effective mass, i.e., heavy fermions. One can conjecture that the observed ESR response of YbRh$_2$Si$_2$ is a novel type of the magnetic resonance excitation given by heavy electrons. Owing to a strong hybridization effect the heavy QPs may inherit 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 spin susceptibility of heavy fermions, χ_p, contributes to the shift of the g factor. This should yield a T dependence similar to that of the electronic specific heat and the NMR Knight shift which is indeed found in YbRh$_2$Si$_2$ (Fig. 2). Compared with the static susceptibility data [6] the g shift shows a crossover to the heavy fermion LFL state, even more clearly suggesting that ESR directly addresses the QPs' spin susceptibility. In fact, the field dependence of the g factor's temperature variation [Figs. 2(a) and 2(c)] reflects what qualitatively would be expected for the development of the heavy fermion state in YbRh$_2$Si$_2$ due to the Kondo effect: with decreasing temperature and/or decreasing magnetic field the Kondo interaction between $4f$ and conduction electron spins enhances, giving rise to heavy fermions, and the g factor deviates from the values of the completely localized $4f$ spin. Remarkably, the ESR signal in YbRh$_2$Si$_2$ becomes observable for *all* magnetic fields at temperatures comparable to or smaller than $T_0 \approx 25$ K, further suggesting that its occurrence is related to the formation of heavy fermions.

In Fig. 3(a) the T dependence of the linewidth was parametrized as $\Delta B(T) = a + bT + c/[\exp(\Delta/T) - 1]$. Here a depicts a residual width, bT stands for the relaxation broadening via electronic degrees of freedom, and the last term has been assigned in Ref. [7] to a relaxation via an excited magnetic state of Yb^{3+} at an energy Δ above the ground state. However, the fit of the HF ESR data requires a significant reduction of Δ from $\sim$115 K (Ref. [7]) to $\sim$50–60 K. Moreover, from neutron scattering results, this excitation energy Δ is much higher ($\Delta \sim 200$ K) [16]. Therefore, the exponential term in $\Delta B(T)$ is not related to the relaxation via excited states. Instead, it seems that the strong broadening of the ESR response mimics the breakdown of the heavy fermion state approaching the T_0/B^* crossover line (Fig. 1).

For $B_{res} \sim 5.15$ and 6.15 T $\Delta B(T) \propto 1/T_2^*(T)$ deviates appreciably from the fit at $T < 7$ K and turns to a T^2 dependence [Fig. 3(b)], suggesting the occurrence of a new spin relaxation regime for heavy fermions. We recall

that this change is concomitant with characteristic crossovers in the ESR $g(T)$ dependence [Fig. 2(a), arrows] as well as in the specific heat (γ) and NMR [K, $1/(T_1T)$] measurables, pointing at the common origin of these features. In particular, the saturation of γ, K, and $1/(T_1T)$ at low temperatures has been associated with the establishment of properties of the LFL state [2,3,5]. Since classically the spin relaxation of QPs is proportional to their momentum relaxation, a T^2 law for $\Delta B(T)$ could be related to the QP-QP scattering in the LFL state. However, in the resistivity the T^2 regime occurs at much lower temperatures [5] (Fig. 1), implying a more complex relationship between the spin and momentum relaxation of heavy electrons in a correlated quantum metal compared to its classical counterpart.

The idea that many-body effects due to strong EE interactions may qualitatively change the ESR response in correlated metals has been discussed already for quite some time. For instance, the occurrence of a narrow collective ESR mode with a considerable shift of the g factor has been predicted in Ref. [13]. Such a sharp mode is expected for an arbitrary sign of the EE interaction, though owing to the enhanced spin susceptibility the ferromagnetic (FM) case is easier to observe experimentally. Striking examples for such a situation are the CESR signals in Pd [17] and TiBe$_2$ [18]. For Kondo lattice systems there is strong experimental evidence that FM correlations between the Kondo ions are essential for narrow observable ESR signals [19]. For YbRh$_2$Si$_2$ in the T-B parameter domain studied in this Letter, FM fluctuations have been found by ^{29}Si-NMR [3]. Very recently, motivated by ESR experiments on YbRh$_2$Si$_2$, two theoretical models of ESR in Kondo lattice systems with anisotropic (Ref. [20]) and isotropic (Ref. [12]) magnetic EE interactions have been proposed. Though using different approaches both theories predict, in particular, for the LFL phase with FM interactions, a sharp ESR line only slightly shifted from the position expected for the local $4f$ resonance. The narrowing takes place by a factor of the mass enhancement m/m^* [12,20] or is due to the action of only the anisotropic part of EE interaction [20]. Remarkably, both predict a T^2 and B^2 dependence of the linewidth in the LFL regime. The former is clearly identified in the data [Figs. 3(a) and 3(b), Fig. 1, left frame]. The latter seems to appear in the dependence on B of the extrapolated residual width at $T = 0$ in Fig. 3(b), at least for the three field values in the LFL regime [21].

In summary, we have studied HF ESR in YbRh$_2$Si$_2$ in a large (T-B) parameter domain extending from the breakdown of the heavy fermion state at large T and large fields until the crossover to the LFL state at lower temperatures. By comparing the T and B dependences of the Sommerfeld coefficient [2,5], the ^{29}Si-NMR Knight shift, and $1/T_1$ rate [3] with those of the ESR g factor and the linewidth ΔB,

the signatures of this crossover have been identified in the ESR measurables. The striking similarity of these dependences strongly suggests that the ESR response is due to a resonance excitation of the quasiparticles in the Kondo lattice, i.e., the "heavy electron spin resonance." As such it thus directly probes the evolution of the Kondo state and the occurrence of different electronic regimes in YbRh$_2$Si$_2$. This conjecture qualitatively explains the magnetic field dependence of (i) the g factor in terms of a gradual change from a T-independent towards a ln$T$-dependent g factor and (ii) the line broadening, in particular, a crossover to a T^2 variation of ΔB at low T in strong fields.

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