Kondo-Ion Electron-Spin Resonance

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One major focus in condensed matter physics is the investigation of compounds where strong correlations among the charge carriers cause unusual, not-understood properties. In this respect, the spin (magnetic) degrees of freedom of the electrons are of particular interest. Here, electron-spin resonance (ESR) spectroscopy allows to investigate elementary magnetic excitations in a standard setup at energies of the order of 0.1 meV.

We performed a detailed ESR study of the strongly correlated electron systems $YbRh_2Si_2$ [1] and $YbIr_2Si_2$ which both exhibit heavy-fermion behavior, i.e., strongly enhanced coefficients of both the electronic specific heat and the T^2 term in the electrical resistivity. These compounds are located very close to a quantum critical point (QCP) corresponding to the disappearance of antiferromagnetic (AF) order (due to the increasing *f*-hybridization) [2, 3]. Low-temperature thermodynamic, transport and magnetic properties suggest that, when approaching the QCP, the heavy quasiparticles seem to disintegrate into a charge part (current) and a spin part (magnetism) [4]. This break-up of heavy quasiparticles was successfully described within a locally critical (LC) scenario [5]. The ESR signal in these compounds clearly shows properties typical of a local Yb^{3+} spin [1]. Obviously, the ESR of the Kondo-ion itself is observed displaying its local character and, therefore, providing experimental evidence for the LC scenario.

There is a considerable amount of literature focussing on ESR investigations of heavy-fermion compounds doped by paramagnetic probes (Gd^{3+} in most cases, see reviews [6, 7]). In the case of $YbRh_2Si_2$, the observed Kondo-ion ESR linewidth is about three orders of magnitude smaller than the linewidth $k_B T_0/\mu_B \cong 37$ T estimated from the spin fluctuation temperature $T_0 \cong 24$ K inferred from thermodynamic measurements [2]. In order to understand the small linewidth and to reveal the underlying mechanism, additional information is desirable and has been found by investigating the ESR in $YbRh_2Si_2$ doped either by Ge on the Si site [8] or by La on the Yb site. Both dopands change the 4 *f*-conduction electron hybridization and hence, the distance to the QCP. In $YbIr_2Si_2$, with $T_0 \cong 40$ K, Ir replaces the smaller Rh resulting in the same effect. The properties of the ESR line are found to be sensitive to this distance [1,8].

Experiment

ESR probes the imaginary part of the dynamic susceptibility Im $\chi(q = 0,\omega)$, and it is sensitive to the local electronic properties of the 4*f* ions. The experimentally probed quantity is the absorbed power P of the sample in a transversal magnetic microwave field (frequencies 9.4 GHz and 34.1 GHz) as a function of an external, static magnetic field B. We used single crystalline platelets of $YbRh_2Si_2$ (pure [1,8] and doped by Ge on the Si site [8] or La on the Yb site [9]) and $YbIr_2Si_2$ [10]. The preparation of these samples as well as their magnetic and transport properties have been described elsewhere [2, 3]. The sample temperature was continuously varied between 0.8 K and 50 K with ^{3}He and ^{4}He cryostats of both flow and bath type.

Results

Figure 1 shows a typical spectrum of $YbRh_2Si_2$ which is recorded as dP/dB vs. B at a frequency of ν = 9.4 GHz. The asymmetry of the Lorentzian-type line shape ("Dysonian") is due to a non-vanishing dispersion contribution to the line and is typical for metallic samples in which the penetration depth is smaller than the sample size, as in our case. From a fit of the experimental spectrum to a Dysonian shape we determined the ESR parameters: resonance field (B_{Res}), linewidth (ΔB), and line intensity (I_{ESR}).

The value of B_{Res} and its angular behavior clearly identify the Yb^{3+} spin as the ESR probe in a tetragonal crystalline symmetry. The inset of Fig. 1 demonstrates the strong anisotropic behavior of the ESR line when the crystal is rotated as shown in the sketch. With $g = h\nu/\mu_B B_{res}$, $g_\perp = 3.561 \pm 0.006$ and $g_\parallel = 0.17 \pm 0.07$ are obtained at $T = 5$ K. These values are typical for the Yb^{3+} (crystal-field derived)

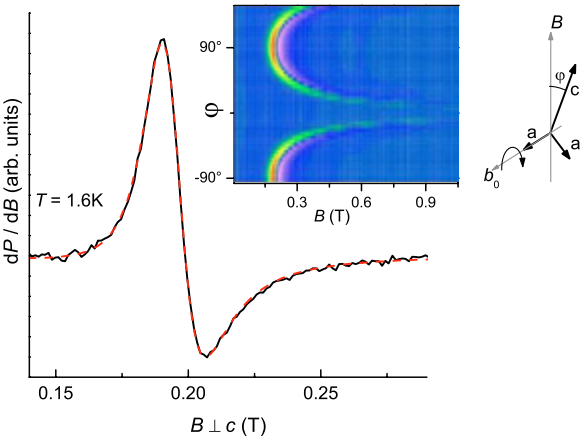

Fig. 1: Representative ESR spectrum of $YbRh_2Si_2$ at T = 1.6 K, i.e., well below the Kondo temperature (25 K). The red dashed line describes the spectrum by a "Dysonian" shape. The inset illustrates the extreme angular dependence of the ESR spectrum observed when rotating the crystal as shown in the sketch (b_0: microwave magnetic field).
This reveals the origin of the resonance, namely local Yb^{3+} spins in a crystalline electric field with tetragonal symmetry.

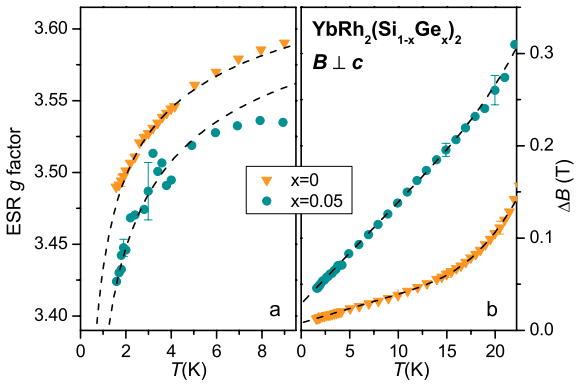

Fig. 2: Comparison of the temperature dependences of ESR line parameters at 9.4 GHz for the pure and Ge-doped compound. (a) Dashed lines describe the data by a g-factor obtained from models valid above the Kondo temperature. (b) Dashed lines represent line width fits with a behavior as observed in conventional Yb-doped intermetallics (details: Refs. [1,8]).

Kramers doublet ground state $^2F_{7/2}$ in tetragonal systems, with possible wave function symmetries Γ_6 or Γ_7 [11], and with close consistency to the large magneto-crystalline anisotropy seen by magnetic susceptibility measurements [2]. Similar results with weaker *g*-value anisotropy are found for $YbIr_2Si_2$ [10].

The ESR intensity I_{ESR} corresponds to the uniform static susceptibility of the ESR probe ions. Typical local moment character is displayed by a Curie-Weiss type temperature dependence of $I_{ESR}(T)$ with a negative Weiss temperature [1, 8, 12]. By comparing $I_{ESR}(T)$ of $YbRh_2Si_2$ with $I_{ESR}(T)$ of $Y_{0.99}Yb_{0.01}Pd_3$ (which has similar B_{Res} and ΔB at 5K) we estimate that at least 60 % of the Yb^{3+} ions contribute to the ESR signal in $YbRh_2Si_2$ [12]. Therefore, the observed ESR indeed is a bulk property. This is an extraordinary result as in dense Kondo lattice systems the ESR of the Kondo ion itself has never been observed *below* the Kondo temperature T_K.

For the *dilute* Kondo system Au:Yb, a Kondo ion (Yb^{3+}) ESR was reported *above* $T_K \approx 0.01$ mK [13]. There, the temperature dependent effective exchange coupling results in a $g(T) \sim \ln (T / \tilde{T}_K)$ dependence. Such a single-ion Kondo scenario consistently describes the low-temperature behavior of $g(T)$ of all our investigated compounds of $YbRh_2Si_2$ as shown in the left frame in Fig. 2 for the pure ($\tilde{T}_K = 20$ mK [1]) and Ge doped compound ($\tilde{T}_K = 35$ mK [8]). Taken as an effective spin-fluctuation (or Kondo) temperature, this value corresponds to a linewidth $\Delta B_K = k_B \tilde{T}_K/\mu_B \cong 30$ mT which is in pretty good agreement with the observed linewidth, see right frame of Fig. 2. The small value of $\tilde{T}_K$ is consistent with the observation of very large unscreened Yb^{3+} moments ($\mu_{eff} \cong 1.4\ \mu_B$) in the $B = 0$ static bulk susceptibility for $T_N < T < 0.3$ K [2]. We suggest that this large discrepancy to the three orders of magnitude larger value of T_K (derived from transport and thermodynamic properties [2, 3]) is related to the local nature of the QCP [5] in these compounds.

The nature of the relaxation mechanism of the Yb^{3+}-spins is reflected by the linewidth ΔB. Its temperature dependence is shown in Figs. 2b and 3 for $B \perp c$ axis. The dashed lines describe the linewidth data as follows: In the region 1 K $\leq T \leq$ 12 K, $\Delta B(T)$ shows an increase linear in temperature which is a behavior typically found for a local moment relaxation in a metallic environment [14]. As shown in Fig. 2b the slope for the Ge-doped sample is larger than that for the undoped sample by about the same factor by which the residual linewidth $\Delta B_0 = \Delta B$ ($T \rightarrow 0$) is increased [8]. This points towards a common relaxation mechanism to which ΔB_0 and the linear term can be ascribed to. The same scaling behavior holds for the La-doped samples [9].

Above $T \cong 12$ K an exponential increase $\Delta B(T) \propto 1/(\exp(\Delta/T) - 1)$ becomes dominant. This is due to a relaxation via an excited energy level Δ above the ground state. The extracted values for Δ are considerably smaller than the first excited crystalline field levels in $YbRh_2Si_2$ and $YbIr_2Si_2$ (found by neutron scattering [15]).

At temperatures below 1 K we observed a deviation from the linear temperature behavior of $\Delta B(T)$ in $YbRh_2Si_2$. This deviation occurs at the crossover temperature boundary which separates Non-Fermi liquid (NFL) behavior from Landau Fermi liquid behavior in the thermodynamic and transport properties [2]. This crossover is indicated at slightly higher temperatures when using the ESR relaxation rates for a comparison with ^{29}Si NMR data [16], shown in the inset of Fig. 3. The nuclear spin relaxation rate $1/T_1$ contains the dynamical $4f$ related susceptibilities according to $(1/T_1T)_{4f} \propto \mathrm{Im}\,\chi(\omega)_{4f}/\omega$; ($\hbar\omega / k_BT \ll 1$). The quantity $(\mathrm{Im}\,\chi/\omega)_{4f}^{ESR}$ includes the temperature dependence of the ESR linewidth, resonance field, and intensity [1]. Both ESR and NMR results show a change of slope at temperatures which depend on the applied magnetic field. At fields $B = 0.5$ T and 2.42 T the saturation originates from a crossover from a NFL regime at elevated temperatures to a field-induced, low-temperature Landau Fermi liquid regime, for which $1/T_1T$ is T-independent [16]. At fields $B \leq$ 0.19 T both, ESR and NMR data, do not show any saturation at low T which was explained with developing critical antiferromagnetic ($q \neq 0$) spin fluctuations when approaching the Néel state [16].

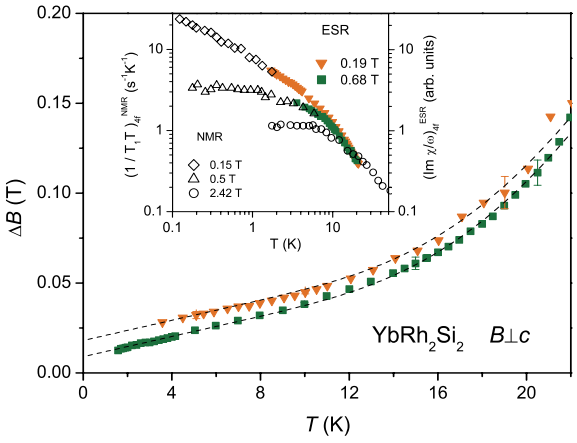

Fig. 3: Temperature dependence of the ESR linewidth ΔB at resonance fields $B_{Res} = 0.19$ T (9.4 GHz, triangles) and 0.68 T (34.1 GHz, squares). Dashed lines fit the data by assuming a behavior observed in conventional Yb-doped intermetallics (details: Ref. [1]). Inset: comparison of spin-lattice relaxation data from ^{29}Si-NMR ([16], open symbols) with data derived from the ESR parameters (closed symbols, details: Ref. [1]).

In order to characterize the spatial dependence of the spin dynamics of the Yb^{3+} spin we investigated the angular dependence of the ESR relaxation rate $\Gamma(\varphi) = \omega_{ESR}\Delta B(\varphi)/B_{Res}(\varphi)$ in the temperature range 4.2 K – 12 K, see Fig. 4. The crystal was rotated as sketched in Fig. 4b with the microwave magnetic field b_0 being always perpendicular to the crystalline c-axis. When keeping φ fixed we found the relaxation rate to be independent on the orientation of the crystalline axes with respect to the microwave magnetic field. By variation of φ the temperature dependence of the line width as shown in Figs. 2b and 3 remains qualitatively unchanged. Quantitatively, a pronounced deviation from the $\Gamma(\varphi = 90°)$-value is visible for $\varphi \leq 30°$. As shown in Figs. 4a and 4b this anisotropy can be attributed to the zero-temperature residual relaxation rate $\Gamma_0(\varphi)$, i.e., $\Gamma - \Gamma_0$ behaves spatially isotropic within our experimental accuracy.

The slope of the T-linear part of $\Delta B(T)$ corresponds to a slope $\partial\Gamma/\partial T$ in the T-linear part of the relaxation rate. The angular variation of $\partial\Gamma/\partial T\,(\varphi)$ is shown in Fig. 4a. $\partial\Gamma/\partial T\,(\varphi)$ appears to be isotropic within the error bars. Within a Fermi-liquid theory, $\partial\Gamma/\partial T \propto [N(E_F)J]^2$ [14]. The isotropic behavior at finite temperatures of the dynamic spin properties of the Kondo ion is consistent with a wave vector independent form of the spin susceptibility within the LC scenario of quantum criticality [5].

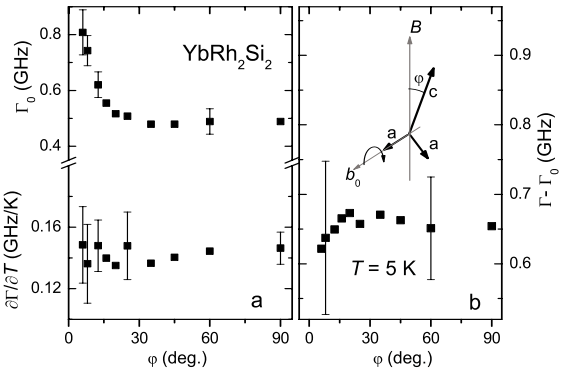

Fig. 4: Angular dependence of the relaxation rate $\Gamma = g\mu_B\Delta B/h$ for the ESR at 9.4 GHz. (a) Angular dependence of both the residual $\Gamma_0 = \Gamma(T=0\ K)$ and the slope $\partial\Gamma/\partial T$ of the T-linear part of $\Gamma(T)$. (b) Angular dependence of the ESR relaxation corrected for the residual relaxation.

Summary and Outlook

Our ESR results on the dense Kondo-lattice systems $YbRh_2Si_2$ and $YbIr_2Si_2$ led to the central conclusion that local magnetic Yb^{3+} moments exist well *below* the characteristic spin fluctuation or Kondo temperature characterizing consistently the thermodynamic and transport properties at higher temperatures. The existence of a well behaved ESR line of Yb^{3+} at such low T indicates an almost complete lack of Kondo screening of the Yb^{3+} magnetic moments. Furthermore, from the field dependence of the ESR relaxation rate (see inset of Fig. 3) as well as from the analysis of the ESR intensity [11] we found evidence of dominating FM fluctuations in $YbRh_2Si_2$ above the critical field, consistent with ^{29}Si NMR results [16]. From our results it is obvious that a simple single-ion Kondo scenario fails to explain our observations. On the other hand, the localized moment scenario for heavy-fermion QCPs implies a type of dynamical susceptibility which relates to local critical degrees of freedom coexisting with spatially extended ones [5]. Such a scenario appears to be strongly supported by our observation that an ESR signal due to local Yb^{3+} moments develops significantly below the ordinary Kondo temperature.

Future work will focus on extending the experimental parameters such that the transition between the non-Fermi liquid and the Landau Fermi-liquid regime is accessible for ESR experiments. Setups for measuring ESR at temperatures down to 0.3 K, with magnetic fields up to 17 T, and at pressures up to 2 GPa are presently under construction. Furthermore, from ESR investigations of Gd-doped $YbRh_2Si_2$ single crystals (which are under preparation at present) promising, additional results concerning the Kondo-ion spin dynamics can be expected. A preliminary theoretical approach has been achieved recently and will be further developed in a future collaboration with Prof. B. I. Kochelaev *et al*. (Kazan State University, Russia).

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