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Electron spin resonance of the Yb 4*f*-moment in Yb(Rh_{1-x}Fe_x)₂Si₂

J. Sichelschmidt^{*1}, H. S. Jeevan², M. Mchalwat², and P. Gegenwart²

¹Max Planck Institute of Chemical Physics of Solids, 01187 Dresden, Germany

²I. Physikalisches Institut, Georg-August Universität, 37077 Göttingen, Germany

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* Corresponding author: e-mail sichelschmidt@cpfs.mpg.de, Phone: +49 351 46463221, Fax: +49 351 46463232

The electron spin resonance (ESR) signal of the heavy-fermion metal YbRh₂Si₂ is investigated in terms of its dependence on the partial substitution of Rh by Fe. This non-isovalent substitution gives rise to a chemical pressure and also leads to a reduction of the conduction electron density of states (hole doping). The values and anisotropy of the ESR parameters are comparable with those of Yb(Rh_{1-x}Co_x)₂Si₂ for Fe-contents up

to at least 10%. This result provides additional evidence that the Kondo interaction allows narrow ESR spectra in a heavy fermion compound. The absence of an ESR signal for 30% Fe content points towards a distribution of different crystal electric field symmetries at the Yb-sites, leading to a dominating disorder-related broadening.

1 Introduction The heavy fermion metal YbRh₂Si₂ has proven to display a variety of unusual low temperature electronic properties which are related to the interplay between the Kondo interaction of Yb³⁺ 4*f* spin/conduction electron spin and the indirect magnetic RKKY interaction of the 4*f* spins. Due to its close vicinity to a magnetic field induced quantum critical point [1] it has been proven as a prototype compound to investigate theoretical scenarios of the Kondo effect breakdown and Fermi surface reconstruction at the antiferromagnetic quantum critical point [2]. Moreover, YbRh₂Si₂ is one of a few Kondo lattice compounds where a well-defined electron spin resonance (ESR) signal allows to directly characterize the spin dynamics of the Kondo ion. Even well below a single ion Kondo temperature ($T_K \simeq 20$ K) [1] this signal shows properties typical for localized 4*f* moments [3, 4]. Various theoretical approaches, invoking the spin-dynamical properties of local and itinerant magnetism in Kondo lattice systems, point out that the origin of the resonance is intimately connected to the Kondo interaction [5–7]. However, compared to T_K , the extraordinary small ESR energy scale is still an open issue to be classified in the physics of Kondo lattice systems. In this respect, it is worth to note that low-temperature ESR investigations of YbRh₂Si₂ have shown the sensitivity of linewidth and *g*-factor to the

characteristic temperature scale T^* [8]. At T^* the Sommerfeld coefficient of the specific heat and the magnetic susceptibility approach a Landau-Fermi liquid behavior [1].

Here we investigate the properties of this ESR signal when substituting the Rh ions with smaller and non-isoelectronic Fe ions, thus reducing the chemical pressure and the electron concentration. This considerably affects the transport properties as shown for the electrical resistance in Fig. 1 for the current within the tetragonal plane. The overall temperature dependence of the resistance in YbRh₂Si₂ is characterized by a broad maximum at T_{\max} . This maximum results from the interplay of crystal electric field (CEF) splitting and the onset of coherence due to the Kondo effect. With increasing Fe-content T_{\max} clearly shifts towards lower temperatures. This shift of T_{\max} and also the residual resistivity ratio is similar to that observed in Yb(Rh_{1-x}Co_x)₂Si₂ [9]. At low *x*, the CEF levels are not influenced much and the shift of T_{\max} could be related to a reduction of the Kondo scale, resulting from the chemical pressure effect [9]. However, there is no direct correspondence of the size of T_{\max} with T_K . The disorder, introduced by the substitution, also effects the onset of coherence. From the fact, that the evolution of $T_{\max}(x)$ for Fe-substitution is very similar to that for Co, we conclude, that in both systems a similar chemical pressure and disorder effect is present, which

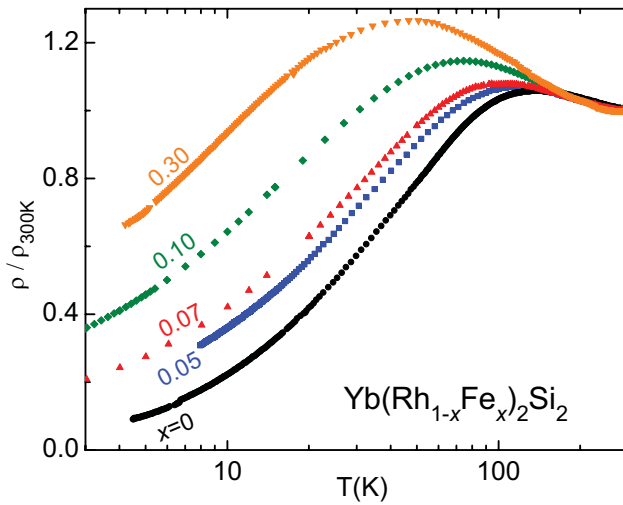


Figure 1 (online colour at: www.pss-b.com) Temperature dependence of the normalized electrical resistance of $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ for different x . With increasing x the characteristic maximum shifts towards lower temperatures and the residual resistivity increases.

dominates the resistivity in this temperature range, although Fe-substitution additionally dopes holes to the system.

2 Experiment A series of single crystalline samples of $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ with $x = 0, 0.05, 0.07, 0.10, 0.3$ was synthesized using an In flux method (for details see Ref. [11]). The Fe-content was checked by characterizing the crystals with scanning electron microscopy equipped with energy dispersive X-ray analysis. We present results of ESR measurements with a standard continuous wave technique at a microwave frequency of $\nu = 9.4 \text{ GHz}$ which according to the resonance condition $h\nu = g\mu_B B_{\text{res}}$ and $g = 3.5$ corresponds to a resonance field of $B_{\text{res}} = 0.19 \text{ T}$. The samples were cooled by a ^4He flow cryostat down to $T = 2.7 \text{ K}$. This temperature is still well above T^* and, therefore, eventual anomalies of the ESR parameters when crossing $T = T^*$ (as seen in YbRh_2Si_2 [8]) could not be investigated. We ensured a proper crystal alignment (external field B within tetragonal a - b -plane; $B \perp c$) by minimizing the highly anisotropic resonance field. All spectra could be nicely fitted with Lorentzian lines (containing dispersive contributions due to a finite penetration depth [12]) as shown by the solid lines in Fig. 2.

3 Results and discussion Figure 2 displays typical ESR spectra (derivative dP/dB of the absorbed microwave power P) for $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ which could be fitted with one Lorentzian line using a constant dispersion to absorption ratio, $D/A = 1$. Eventual resonance contributions from spurious Fe-containing phases larger than 0.01% could not be detected. We thus exclude Fe-ions as a possible resonance source. This is also supported by the obtained fitting quality with one single line showing parameters comparable to those of Co-containing YbRh_2Si_2 [10].

For the concentration $x = 0.3$ no ESR signal could be detected. We suspect that such an amount of Fe-substitution

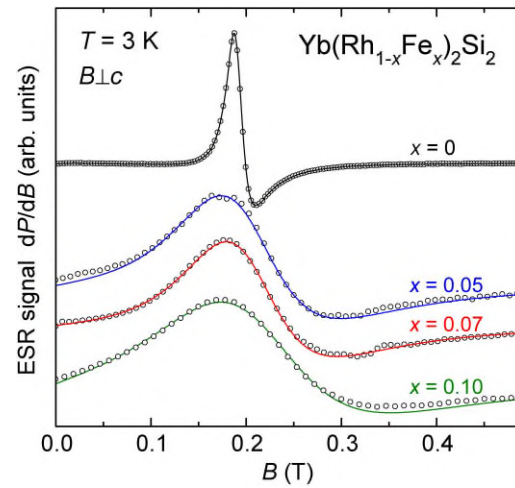


Figure 2 (online colour at: www.pss-b.com) Selected ESR spectra of $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ at $T = 3 \text{ K}$ with c -axis perpendicular to the applied magnetic field B . Solid lines indicate Lorentzian line fits with same ratios of dispersion to absorption.

creates a dominating inhomogeneous line-broadening from a considerable disorder in the crystal-field distribution. In contrast, the disorder effect due to 30% *isoelectronic* Co-substitution is much smaller, leaving the ESR line well observable [10]. For all other investigated Fe-contents of $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ the ESR spectra display strong uniaxial anisotropy in their resonance field (see inset of Fig. 3) and linewidth regarding the rotation of the crystalline c -axis in the external magnetic field B . The corresponding g -values g_{\parallel} and g_{\perp} for $B \parallel c$ and $B \perp c$, respectively, are shown in Fig. 3. These values are expected for $\text{Yb}^{3+} 4f$ -ions in the crystal field of YbRh_2Si_2 [13]. Comparing the anisotropy g_{\perp}/g_{\parallel} of Fe- and Co-substituted YbRh_2Si_2 we observe a $\approx 50\%$ reduced

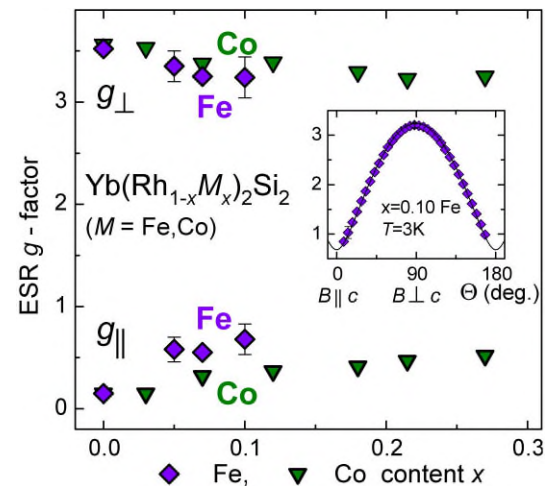


Figure 3 (online colour at: www.pss-b.com) Dependence of the ESR g -factor at $T = 5 \text{ K}$ on the Fe content in $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ (diamonds) and on the Co content in $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ (triangles, data from Ref. [10]). Inset shows the uniaxial anisotropy (solid line) of the g -values for $x = 0.10$ Fe content.

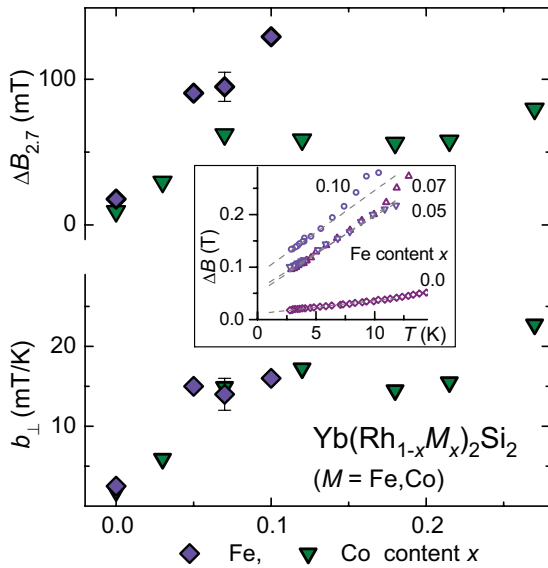


Figure 4 (online colour at: www.pss-b.com) Dependence of ESR linewidth $\Delta B_{2,7}$ at $T = 2.7$ K and linewidth temperature slope b_{\perp} on the Fe content in $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ (squares, diamonds) and on the Co content in $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ (triangles, data from Ref. [10]). Inset shows temperature dependence of linewidth ΔB ($B \perp c$) with linear behavior (dashed lines) providing $\Delta B_{2,7}$ and b_{\perp} .

anisotropy for an Fe-substitution of $x = 0.10$. We relate this reduction to differences in the charge distribution at the Rh lattice sites.

The temperature dependence of the ESR g -factors shows the same behavior as those of the corresponding Co-concentration in YbRh_2Si_2 . There, as shown in Fig. 6 of Ref. [10], a characteristic logarithmic downturn towards low temperatures was observed. We refrain from a quantitative comparison of the g -factor temperature dependence of Fe- and Co-substituted YbRh_2Si_2 because of a 5% error due to the fitting of rather broad lines.

The spin-dynamics as reflected in the ESR linewidth shows a temperature and concentration dependence which is typically observed in YbRh_2Si_2 [10]. As shown in the inset of Fig. 4, the data could be described by a Korringa-like increase linear in temperature up to about 10 K. For $x = 0$, at higher temperatures, an exponential increase can be resolved which is due to spin-phonon interaction caused by crystal field modulations via lattice vibrations [14].

The main frame of Fig. 4 shows the parameters of the linear temperature dependence of the linewidth $\Delta B_{2,7}$ at $T = 2.7$ K and linewidth temperature slope b_{\perp} for $B \perp c$. For $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ $\Delta B_{2,7}$ was shown to correlate with the reciprocal characteristic temperature $1/T_{4f}$ which reflects the energy scale of all exchange interactions acting on the lowest crystalline electric field doublet [10]. This suggested that a decrease of the Kondo energy scale T_K leads to an increase of the ESR linewidth. This relation between ΔB and T_K was also shown to hold for the slope b for up to $x \approx 0.2$. The Fe/Co comparison of $\Delta B_{2,7}$ and b_{\perp} shown in Fig. 4 demonstrates the same trend and, hence, the Kondo

interaction controls the linewidth in both Fe- and Co-substituted YbRh_2Si_2 . However, the clearly larger values of $\Delta B_{2,7}$ for $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$ point towards the influence of the disordered distribution of different CEF symmetries which is considerably stronger if Rh-sites are substituted by non-isoelectronic Fe-ions. We suspect that for the same reason the ESR line for $x = 0.3$ is not observable.

4 Conclusions The local magnetic properties of the Kondo ion Yb^{3+} could clearly be characterized by the ESR of $\text{Yb}(\text{Rh}_{1-x}\text{Fe}_x)_2\text{Si}_2$. Similar to the application of pressure or isoelectronic Co-substitution the behavior of the ESR parameters indicate the effect of the Kondo interaction between $4f$ spins and conduction electrons. Differences in the ESR parameters between Fe- and Co-substituted YbRh_2Si_2 are mainly seen in a reduced anisotropy of the g -factors and in an enhanced disorder-broadening for larger Fe-contents.

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