On the local and itinerant properties of the ESR in YbRh$_2$Si$_2$

Jan Wykhoff$^a$, Jörg Sichelschmidt$^{a,*}$, Gerard Lapertot$^b$, Georg Knebel$^b$, Jacques Flouquet$^b$, Ilshat I. Fazlishanov$^d$, Hans-Albrecht Krug von Nidda$^c$, Cornelius Krellner$^a$, Christoph Geibel$^a$, Frank Steglich$^a$

$^a$Max-Planck-Institut für Chemische Physik fester Stoffe, 01187 Dresden, Germany
$^b$Département de la Recherche Fondamentale sur la Matière Condensée, SPSMS, CEA Grenoble, 38054 Grenoble, France
$^c$Experimentalphysik V, EKM, Universität Augsburg, 86135 Augsburg, Germany
$^d$E.K. Zavoisky Physical-Technical Institute, 420049 Kazan, Russia

Received 25 May 2007; received in revised form 13 July 2007; accepted 20 July 2007
Available online 27 August 2007

Abstract

Below the Kondo temperature the heavy Fermion compound YbRh$_2$Si$_2$ shows a well defined electron spin resonance (ESR) signal which features properties similarly observed for local Yb$^{3+}$ ions in metallic environments [1]. Remarkably, this signal was observed at resonance energies much smaller than the Kondo energy and well below the Kondo temperature $T_K \approx 25$ K, where the magnetic Yb$^{3+}$ moments are supposed to be largely screened and ESR silent. This seemingly contradiction is an important, yet unexplained feature of the magnetism in YbRh$_2$Si$_2$. This compound is located very close to a magnetic field induced quantum critical point where weak antiferromagnetic (AF) ordering is suppressed in a field of 70 mT applied in the magnetic easy tetragonal $ab$ plane [2]. Besides the AF order at $T_N = 70$ mK pronounced ferromagnetic fluctuations are evident from a highly enhanced Sommerfeld–Wilson ratio [3] and from the spin dynamics as seen in $^{29}$Si NMR measurements [4].

The existence of the ESR in YbRh$_2$Si$_2$ below $T_K$ seems even more surprising when its local character is envisaged: besides its Weiss-like temperature dependence of the intensity it shows a pronounced anisotropy which perfectly agrees with the magnetocrystalline anisotropy [5]. However, $^{170}$Yb Mössbauer spectra indicate a much faster Yb$^{3+}$ relaxation than one would infer from the ESR linewidth [10]. On the other hand, assuming the local moments to be Kondo screened, conduction carriers themselves could cause a narrow and intense ESR signal because strong ferromagnetic fluctuations are present. This kind of conduction ESR is reported, for instance, in Pd [6] or in TiBe$_2$ [7].

With this paper we highlight how both scenarios (local/itinerant) may characterize the ESR spectra. For this purpose we present (i) an estimation of the ESR intensity which is involved in the YbRh$_2$Si$_2$ ESR compared to the local Yb$^{3+}$ ESR of YPd$_3$:Yb and (ii) a discussion of the lineshape’s itinerant character which may be seen if spin

1. Introduction

The heavy fermion compound YbRh$_2$Si$_2$ shows a well defined electron spin resonance (ESR) signal which features properties similarly observed for local Yb$^{3+}$ ions in metallic environments [1]. Remarkably, this signal was observed at resonance energies much smaller than the Kondo energy and well below the Kondo temperature $T_K \approx 25$ K, where the magnetic Yb$^{3+}$ moments are supposed to be largely screened and ESR silent. This seemingly contradiction is an important, yet unexplained feature of the magnetism in YbRh$_2$Si$_2$. This compound is located very close to a magnetic field induced quantum critical point where weak antiferromagnetic (AF) ordering is suppressed in a field of 70 mT applied in the magnetic easy tetragonal $ab$ plane [2]. Besides the AF order at $T_N = 70$ mK pronounced ferromagnetic fluctuations are evident from a highly enhanced Sommerfeld–Wilson ratio [3] and from the spin dynamics as seen in $^{29}$Si NMR measurements [4].

The existence of the ESR in YbRh$_2$Si$_2$ below $T_K$ seems even more surprising when its local character is envisaged: besides its Weiss-like temperature dependence of the intensity it shows a pronounced anisotropy which perfectly agrees with the magnetocrystalline anisotropy [5]. However, $^{170}$Yb Mössbauer spectra indicate a much faster Yb$^{3+}$ relaxation than one would infer from the ESR linewidth [10]. On the other hand, assuming the local moments to be Kondo screened, conduction carriers themselves could cause a narrow and intense ESR signal because strong ferromagnetic fluctuations are present. This kind of conduction ESR is reported, for instance, in Pd [6] or in TiBe$_2$ [7].

With this paper we highlight how both scenarios (local/itinerant) may characterize the ESR spectra. For this purpose we present (i) an estimation of the ESR intensity which is involved in the YbRh$_2$Si$_2$ ESR compared to the local Yb$^{3+}$ ESR of YPd$_3$:Yb and (ii) a discussion of the lineshape’s itinerant character which may be seen if spin
twenty. The ESR linewidth is well characterized by a contribution from unresolved hyperfine lines.

2. Experimental details

ESR probes the absorbed power \( P \) of a transversal magnetic microwave field as a function of an external, static magnetic field \( B \). To improve the signal-to-noise ratio, a lock-in technique is used by modulating the static field, which yields the derivative of the resonance signal \( dP/dB \). The ESR experiments were performed at X-band frequencies (\( v \approx 9.4 \text{ GHz} \)) with a Bruker Elexsys 500 spectrometer. The temperature was varied between 2.7 \( K \leq T \leq 300 \text{ K} \) by using a He-flow cryostat. For the ESR-measurements down to the lowest accessible temperature of \( T = 0.69 \text{ K} \) a homebuilt \(^3\)He cold-finger bath cryostat was used.

We used high quality single crystalline platelets of \( \text{YbRh}_2\text{Si}_2 \) and \(^{174}\text{YbRh}_2\text{Si}_2 \) with small residual resistivities as low as \( \rho_0 = 0.5 \mu \Omega \text{ cm} \) and with very sharp anomalies in the specific heat at \( T = T_N \). Their preparation as well as their magnetic and transport properties were thoroughly described: \( \text{YbRh}_2\text{Si}_2 \) in Refs. [2,8] and \(^{174}\text{YbRh}_2\text{Si}_2 \) in Refs. [9,10]. The crystals were mounted in the microwave cavity such that the microwave magnetic field was always within the tetragonal basal plane.

The polycrystalline bulk sample of \( \text{YPd}_3: \text{Yb} \) was prepared by argon-arc-melting stoichiometric amounts of \( \text{Y}, \text{Pd} \) and dopant amounts of \( \text{YbPd}_3 \) [11]. The latter was prepared before in an induction furnace because the usage of pure \( \text{Yb} \) metal would lead to a considerable \( \text{Yb} \) loss due to its high vapour pressure. Debye–Scherrer X-ray diffraction confirmed a single-phase sample. A \( \text{Yb} \) concentration of 0.6\% was determined by SQUID magnetization measurements.

3. Results and discussion

We first address the question how much intensity is involved in the \( \text{YbRh}_2\text{Si}_2 \)–ESR compared to the ESR of a well localized \( \text{Yb} \) ion, \( \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3 \), \( x = 0.6\% \). The latter compound is suitable for this comparison because it displays ESR properties typical of a local \( \text{Yb}^{3+} \) magnetic moment. The ESR linewidth is well characterized by a Korringa (temperature linear) relaxation and a first excited \( (T_2) \) level at \( \approx 50 \text{ K} \). The temperature independent effective \( g \)-factor of 3.34 is compatible with a doublet \( \Gamma_7 \) ground-state [11,12].

The main frame of Fig. 1 shows the ESR signals of both above mentioned compounds, recorded at \( T = 4.3 \text{ K} \) with the same experimental conditions. Note that Fig. 1 shows the ESR signal amplitudes after normalization to the amount of \( \text{Yb} \) ions by proper consideration of the molar volume which is probed by the microwave in the skin depth \( (\approx 1.3 \mu \text{ m} \text{ for } \text{YbRh}_2\text{Si}_2, \approx 2.3 \mu \text{ m} \text{ for } \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3) \). From simply inspecting Fig. 1 it is already clear at first sight that the ESR of \( \text{YbRh}_2\text{Si}_2 \) cannot be caused by a few percent of local \( \text{Yb}^{3+} \) moments. It is even obvious that the previously assessed lower bound of 60\% ESR active \( \text{Yb} \) ions [13] is underestimated. With respect to a more accurate statement we compare in the following the bulk magnetic susceptibility with the ESR intensity which is proportional to the spin susceptibility of the ESR probe. In general, the ESR intensity can be determined by the area \( \propto \text{Amp} \cdot \Delta B^2 \cdot \sqrt{\Delta \chi^2 + 1} \) under the ESR absorption signal. Here \( \text{Amp} \) is the amplitude, \( \Delta B \) the linewidth, and \( \chi \) the susceptibility parameter (describing the dispersion contribution in metallic samples) of an ESR line with a Lorentzian shape:

\[
\frac{dP}{dB}(B) = \text{Amp} \cdot \frac{\chi (1 - x^2) - 2 \cdot x}{(1 + x^2)^2} + \frac{-\chi (1 - y^2) - 2 \cdot y}{(1 + y^2)^2}
\]

\[
+ a \cdot B + b.
\]

Here \( x = (B - B_0)/\Delta B \), \( y = (B + B_0)/\Delta B \), \( B_0 \) is the resonance field, and \( a \cdot B + b \) denotes the linear background. The second term describes the influence of the counter rotating component of the linearly polarized microwave field which is relevant in the case of \( \Delta B \gg B_0 \). Using the \( \text{Yb} \)-normalized spectra as shown in Fig. 1 we fitted \( \text{YbRh}_2\text{Si}_2 \) with one metallic Lorentzian and \( \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3 \), \( x = 0.6\% \) with one central line caused by the even isotopes \( \text{Yb}^{3+} \) with nuclear spin \( I = 0 \) and eight hyperfine split lines caused by the odd isotopes \( ^{171}\text{Yb}^{3+} \) \( (I = \frac{3}{2}) \) and \( ^{173}\text{Yb}^{3+} \) \( (I = \frac{5}{2}) \). For the latter we employed a third order hyperfine splitting formula with hyperfine constants according to Ref. [11] and we forced \( \Delta B \) and \( \chi \) to be the same for all lines. As shown by the dashed line in Fig. 1 this procedure yielded a quite satisfying fit for \( \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3 \), \( x = 0.6\% \). Moreover, the ratio of the intensities of the central line and the hyperfine split lines

![Fig. 1. ESR signals (dP/dB) of \( \text{YbRh}_2\text{Si}_2 \) (single crystal, \( B \perp \text{c} \)) and \( \text{Yb}^{3+} \) in \( \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3 \) (polycrystal) at 9.4 GHz and \( T = 4.3 \text{ K} \) normalized to the \( \text{Yb} \) content. Dashed lines denote fit curves: one metallic Lorentzian shape for \( \text{YbRh}_2\text{Si}_2 \) and nine metallic Lorentzians with hyperfine structures for \( \text{Y}_{1-x}\text{Yb}_x\text{Pd}_3 \) according to Ref. [11] as described in the main text. The \( \text{YbRh}_2\text{Si}_2 \) data correspond to a sample batch with the lowest residual ESR linewidth. Inset: magnetic susceptibility \( \chi_{\text{Yb}} \) normalized to the \( \text{Yb} \) content and measured at the ESR resonance field \( B = 0.19 \text{ T} \).](image-url)
YbRh$_2$Si$_2$ is shown in Fig. 2 and the ESR spectra are
approximately corresponds to the natural abundance ratio
of even and odd Yb isotopes.

Comparing the fit results for both compounds (the
dashed lines in Fig. 1) we found that their ESR intensities
have a ratio of 1.0 $\pm$ 0.3. This is a remarkable result
because it is not expected from the ratio of their bulk
magnetic susceptibilities per Yb, $\chi_{\text{Yb}}$, as can be seen in the
inset of Fig. 1. $\chi_{\text{Yb}}$ of Y$_1$-xYb$_x$Pd$_3$, $x = 0.6$% considerably
exceeds $\chi_{\text{Yb}}$ of the partly Kondo screened Yb$^{3+}$
magnetic moments in YbRh$_2$Si$_2$, although our comparison with a
local Yb$^{3+}$ ESR probe clearly demonstrates that the full
magnetic moment of the Yb ions in YbRh$_2$Si$_2$ contributes
to the observed ESR signal. At present theoretical
treatments are not available and the reason for this
discrepancy remains unclear. We suspect that the strong
interaction between 4f spins and conduction electron spins
leads to a total spin dynamics which can be observed by
ESR.

Next, we check the assumption that unscreened, local
Yb$^{3+}$ moments could cause an ESR signal with a
characteristic energy which is much smaller than the
Kondo temperature. In this case a hyperfine coupling
between the 4f electron and the Yb nuclear spin should
contribute to the ESR spectra. $^{171}$Yb and $^{173}$Yb nuclei may
provide hyperfine energies near 1 K [10]. We therefore
investigated the ESR of mono-isotopic $^{174}$YbRh$_2$Si$_2$. The
isotope $^{174}$Yb has zero nuclear spin and hence no hyperfine
interactions could be involved in a putative Yb$^{3+}$ ESR in
$^{174}$YbRh$_2$Si$_2$. The temperature behavior of ESR $g$-value
and linewidth $\Delta B$ of mono-isotopic and mixed-isotopic
YbRh$_2$Si$_2$ is shown in Fig. 2 and the ESR spectra are
plotted in Fig. 3b for a representative temperature. $\Delta B(T)$
reflects a typical ESR behavior of a magnetic moment in a
metallic host [1,14]. Both data sets in Fig. 2, $g$-value
and linewidth $\Delta B$, coincide well within the experimental error in
the complete accessed temperature range. Therefore,
hyperfine interactions obviously do not contribute to the
ESR relaxation and resonance field in YbRh$_2$Si$_2$. As will be
shown below, the lineshape also does not yield indications
for hyperfine contributions. Moreover, the ESR intensity
per Yb ion is the same in $^{174}$YbRh$_2$Si$_2$ and YbRh$_2$Si$_2$. We
point out that for the data in Fig. 2 we used two samples
with similar linewidth and similar residual resistivity ratio.
As reported earlier [14] the linewidth at a given tempera-
ture is related to the residual resistivity ratio which in turn
determines the residual linewidth $\Delta B_0$ (temperature-linear
part of $\Delta B$ extrapolated to zero temperature). The
temperature dependence of the linewidth obeys the scaling
$\Delta B^* = (\Delta B - \Delta B_0)/\Delta B_0$, i.e. $\Delta B^* (T)$ for all investigated
YbRh$_2$Si$_2$, for La-doped YbRh$_2$Si$_2$ [14] and also for
$^{174}$YbRh$_2$Si$_2$ collapse onto one single curve. This scaling
provides evidence that the lattice relaxation of a strongly
coupled Yb$^{3+}$-conduction electron system is an impo-
tant ingredient for understanding the ESR linewidth in
YbRh$_2$Si$_2$. This situation resembles the bottleneck-relaxation
mechanism which was discussed extensively for diluted
magnetic moments in metallic hosts [15].

From a putative ESR of a strongly coupled Yb$^{3+}$
conduction electron system one would expect typical
features of an itinerant ESR probe. For example the
lineshape should be influenced by the ESR probe spin
diffusion as theoretically described by Dyson [16] and
resolved hyperfine line-splittings in the ESR of YbRh$_2$Si$_2$.

The deviations from a Lorentzian lineshape arise from nonzero nuclear spin moment and, hence, are reported for the isotope $^{171}$Yb diluted in a Au metallic matrix [18]. In dense systems like YbRh$_2$Si$_2$ such line splittings might be completely absent due to an exchange narrowing process [15, Section 3.2].

4. Conclusion

We compared the ESR of YbRh$_2$Si$_2$ with the ESR of a system which contains a well defined, local Yb$^{3+}$ moment, $Y_{1-x}$Yb$_x$Pd$_3$, $x = 0.6\%$. From relating the ESR intensities of these compounds to their Yb content and taking into account their difference in magnetic susceptibility we conclude that the ESR of YbRh$_2$Si$_2$ involves the magnetic moments of all its Yb ions.

Assuming an itinerant ESR probe in YbRh$_2$Si$_2$ a corresponding lineshape description within the theory of Dyson [16] yields a spin diffusion time in the infinite limit.

However, such a description still leaves small systematic differences between lineshape fit and data which cannot be explained by unresolved hyperfine structures. We suspect that these differences are related to the ESR spin dynamics of the strongly coupled Yb$^{3+}$-conduction electron system in YbRh$_2$Si$_2$.

Acknowledgement

The low-temperature ESR-measurements at the University of Augsburg were partially supported by the German BMBF under Contract no. VDI/EKM13N6917 and by the Deutsche Forschungsgemeinschaft within SFB484 (Augsburg). I.I. Fazlishanov acknowledges support by the Volkswagen Foundation (I/82203).

References

[1] J. Sichelschmidt, V. Ivanshin, J. Ferstl, C. Geibel, F. Steglich, Phys. Rev. Lett. 91 (2003) 156401.
[2] O. Trovarelli, C. Geibel, S. Mederle, C. Langhammer, F.M. Grosh, P. Gegenwart, M. Lang, G. Sparn, F. Steglich, Phys. Rev. Lett. 85 (2000) 626.
[3] P. Gegenwart, J. Custers, Y. Tokiwa, C. Geibel, F. Steglich, Phys. Rev. Lett. 94 (2005) 076402.
[4] K. Ishida, K. Okamoto, Y. Kawasaki, Y. Kitaoka, O. Trovarelli, C. Geibel, F. Steglich, Phys. Rev. Lett. 89 (2002) 107202.
[5] J. Sichelschmidt, J. Wykhoff, H.-A. Krug von Nidda, J. Ferstl, C. Geibel, F. Steglich, J. Phys. Condens. Matter 19 (2007) 116204.
[6] P. Monod, J. Phys. Colloq. C 6 (Suppl. 8) (1978) 1472.
[7] D. Shahtiel, D. Joshe, A. Grayevsky, V. Zevin, J.L. Smith, Phys. Rev. B 36 (1987) 4090.
[8] P. Gegenwart, J. Custers, C. Geibel, K. Neumaier, T. Tayama, K. Tenya, O. Trovarelli, F. Steglich, Phys. Rev. Lett. 89 (2002) 056402.
[9] G. Knebel, V. Glazkov, A. Pourret, P.G. Niklowitz, G. Lapertot, B. Salce, J. Flouquet, Physica B 359 (2005) 20.
[10] G. Knebel, R. Bourisier, E. Hassinger, G. Lapertot, P.G. Niklowitz, A. Pourret, B. Salce, J.P. Sanchez, I. Sheikin, P. Bonville, H. Harima, J. Flouquet, J. Phys. Soc. Japan 75 (2006) 114709.
[11] T. Gambke, B. Eilschener, R. Kremer, M. Schanz, J. Magn. Magn. Mater. 36 (1983) 115.
[12] C. Rettori, E. Weber, J.P. Donoso, F.C.G. Gandra, G.E. Barberis, Physica B+C 107 (1981) 359.
[13] J. Sichelschmidt, V.A. Ivanshin, J. Ferstl, C. Geibel, F. Steglich, J. Magn. Magn. Mater. 272–276 (2004) 42.
[14] J. Wykhoff, J. Sichelschmidt, J. Ferstl, C. Krellner, C. Geibel, F. Steglich, I. Fazlishanov, H.-A. Krug von Nidda, Physica C 2007, doi:10.1016/j.physc.2007.03.123.
[15] S. Barns, Adv. Phys. 20 (1981) 801.
[16] F. Dyson, Phys. Rev. 98 (1955) 349.
[17] G. Feher, A.F. Kip, Phys. Rev. 98 (1955) 337.
[18] Y. von Spalden, E. Tsang, K. Baberschke, P. Schlottmann, Phys. Rev. B 28 (1983) 24.