Field-dependent collective ESR mode in YbRh$_2$Si$_2$

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ABSTRACT

Electron spin resonance (ESR) experiments in YbRh$_2$Si$_2$ Kondo lattice ($T_K \approx 25$ K) at different field/frequencies (4.1 \leq \nu \leq 34.4$ GHz) and $H_{ex}$ revealed: (i) a strong field dependent $Yb^{3+}$ spin–lattice relaxation, (ii) a weak field and $T$-dependent effective $g$-value, (iii) a suppression of the ESR intensity beyond 15\% of Lu-doping, and (iv) a strong sample and Lu-doping $\%$ dependence of the ESR data. These results suggest that the ESR signal in YbRh$_2$Si$_2$ may be due to a coupled $Yb^{3+}$–conduction electron resonant collective mode with a subtle field-dependent spins dynamic.

1. Introduction

The heavy-fermion (HF) Kondo lattice YbRh$_2$Si$_2$ ($T_K \approx 25$ K) is an antiferromagnetic (AF, $T_N = 70$ mK) tetragonal ([4/3]mm) intermetallic compound. At low-$T$ ($T \leq T_K$) the magnetic susceptibility exhibits a HF behavior and at high-$T$ ($T > 200$ K) an anisotropic Curie–Weiss with a full Yb$^{3+}$ magnetic moment ($\mu_{eff} \approx 4.5\mu_B$) is observed [1–3]. The AF ordering of YbRh$_2$Si$_2$ may be driven to $T_N \approx 0$ by fields of $H_{ex} \approx 650$ Oe and $H_{ex} \approx 7$ kOe [4]. At these fields a quantum critical point (QCP) is observed with non-Fermi-liquid (NFL) behavior [1,2]. Therefore, among other systems [5,6], YbRh$_2$Si$_2$ is particularly an interesting system to study quantum criticality and NFL behavior.

Electron spin resonance (ESR) experiments at low-$T$ ($T \leq 20$ K) in YbRh$_2$Si$_2$ by Sichelschmidt et al. [7] have reported on a narrow ($100 – 200$ Oe) single dysonian resonance with no hyperfine components, $T$-dependence of the linewidth, $\Delta H$, and a $g$-value anisotropy consistent with Yb$^{3+}$ in a metallic host of tetragonal symmetry. As in the early work of Tien et al. [8] the observation of a narrow Yb$^{3+}$ ESR in the intermediate valence compound of YbCuAl and, recently, in a dense Kondo system below $T_K$ were unexpected results [7]. Nevertheless, various reports were already published on the ESR of Yb$^{3+}$ in stoichiometric YbRh$_2$Si$_2$ [9], Yblr$_2$Si$_2$ [10], and YbRh$_2$Si$_2$ doped with non-magnetic impurities as Ge [11] and La [12]. Also, the ESR of Ce$^{3+}$ in dense Kondo systems was communicated [13]. However, it is not clear yet what mechanism allows the observation of the Yb$^{3+}$ ESR line with local magnetic moment features in these highly correlated electron systems.

The main purpose of this work is to investigate the $H$-dependent ESR data in the NFL phase of YbRh$_2$Si$_2$ ($4.2 \leq H \leq 10$ K; $0 < H < 10$ kOe). We found an unexpected $H$-dependence behavior of the Yb$^{3+}$ ESR data in YbRh$_2$Si$_2$ that, we hope, will contribute to the understanding of the observed ESR signal in this system.

2. Experiment

Single crystals of Yb$_{1-x}$Lu$_x$Rh$_2$Si$_2$ ($0 \leq x \leq 1.00$) were grown from In and Zn-fluxes as reported elsewhere [14–16]. The structure and phase purity were checked by X-ray powder diffraction. The high quality of our undoped crystals was confirmed by X-rays rocking curves which revealed a mosaic structure of maximum c-axis angular spread of $\approx 0.015$. The electrical residual resistivity ratio, $\rho_{300K}/\rho_{1.9K}$, for the In and Zn-flux grown crystals were 35 and 10, respectively [14–16]. For the ESR spectra $\approx 2 \times 2 \times 0.5$ mm$^3$ single crystals were used. The ESR experiments were carried out in a Bruker S, X, and Q-bands (4.1, 9.5 and 33.8 GHz) spectrometer using appropriated...
resonators and T-controller systems. A single anisotropic resonance, with no hyperfine components, from the Kramer doublet ground state was observed at all bands. The dysonian lineshape \(A/B \approx 2.5\) corresponds to a microwave skin depth smaller than the size of the crystals [17].

3. Results and discussions

Fig. 1 shows the YbRh₂Si₂ ESR X-band spectra at 4.2 K and \(H_{ce}\) for single crystals grown in In and Zn-fluxes. From the anisotropy of the field for resonance, \(H_{0}(T)\) (not shown), one can obtain the angular-dependence of the effective \(g\)-value which is given by \(hν/μ_B H_{o}(θ) = g(θ) = [g_\perp^2, \cos^2 θ + g_\parallel^2, \sin^2 θ]^{1/2}\). From the fitting of the experimental \(H_{0}(T)\) we obtain \(g_\perp \approx 0.3(4)\) and \(g_\parallel = 3.6(0.7)\). The inset of Fig. 1 displays, for \(H_{ce}\), the Korringa-like [18,19] linear thermal broadening of the linewidth, \(ΔH(T) = a + bT\), and the fitting parameters for the crystal grown in Zn-flux at X and Q-bands. As it will be shown below, the large values measured for \(a\) and \(b\) indicates that Zn impurities were incorporated in this crystal.

Fig. 2 presents the relative normalized X-band integrated ESR intensities for Yb₁₋ₓLuₓRh₂Si₂ at 4.2 K and \(H_{ce}\) as a function of \(x\), \(I_{4.2}(x)/I_{4.2}(0)\). The ESR intensities were determined taking into consideration the crystal exposed area, skin depth and spectrometer conditions. These results show that, while for \(x ≤ 0.15\) the Yb\(^{3+}\) ESR intensity is nearly constant, for \(0.15 < x ≤ 1.00\) the intensity vanish completely and no ESR could be detected. It is worth mention that for \(x > 0.15\) and \(T > 200\) K, \(χ_{ce}(T)\) follows an Curie–Weiss law with a full Yb\(^{3+}\) magnetic moment and that for \(x ≤ 0.15\) there is no appreciable changes in the thermodynamic properties of these compounds [14,15]. The absence of resonance for \(x > 0.15\) strongly suggests that the observed ESR for \(x < 0.15\) cannot be associated to a single Yb\(^{3+}\) ion resonance but rather to a resonant collective mode of exchange coupled Yb\(^{3+}\)–ce (conduction electrons) magnetic moments. We argue that a strong Yb\(^{3+}\)–ce exchange coupling may broadens and shifts the ce resonance toward the Yb\(^{3+}\) resonance allowing their overlap and building up a Yb\(^{3+}\)–ce coupled mode with possibly bottleneck/dynamic-like features. Evidences for bottleneck/dynamic-like features in the Lu-doped crystals will be published elsewhere. An internal field caused by the Yb\(^{3+}\) local moments may be responsible for the shift of the ce resonance [21]. Moreover, the Lu-doping may disrupt the collective mode coherence and, probably, may also opens the bottleneck/dynamic regime [22].

Figs. 3a and b show, respectively, the low-T dependence of \(ΔH\) and effective \(g\)-value of the Yb\(^{3+}\) ESR in In-flux grown YbRh₂Si₂.
measured at S, X and Q-bands for \( H_{\perp} \). In this T-interval, and within the error bars, it is also found that \( \Delta H = a + bT \) for the three bands. This suggests a Korringa-type of mechanism for the Yb\(^{3+}\) spin–lattice relaxation (SLR), i.e. the Yb\(^{3+}\) local moment is exchange coupled to the conduction electrons [18]. The residual linewidth, \( a \), and relaxation-rate, \( b = \Delta H/\Delta T \), are given in Fig. 3a. Notice that the minimum relaxation rate, \( b \), is found at X-band, \( H \approx 1900\) Oe. The actual determination of the residual linewidth, \( a = \Delta H(T=0) \), would require measurements at lower-T, therefore, the obtained values should be considered just as fitting parameters. A \( H \)-dependent SLR-rate \( b \) is not expected for a normal local magnetic moment–ce exchange coupled system, where the Korringa-rate is frequency/field independent [19]. However, since the Yb\(^{3+}\) and ce magnetic moments carry unlike spins and different g-values, the \( H \)-dependence of \( b \) may be an anomalous manifestation of a bottleneck-like behavior. Fig. 3b shows that the \( T \)-dependence of the effective \( g \)-values are slightly different in the three bands, with minimum effective \( g_{\perp2} \)-values also at the X-band. The effective \( g \)-value accuracy is much higher than that obtained from \( \hbar/\mu_B H_{\perp}(\theta) = g(\theta) = \left[ g^2_{\perp2} \cos^2 \theta + g^2_{\perp0} \sin^2 \theta \right]^{1/2} \) because proper experimental conditions were chosen for these \( H_{\perp} \) measurements. 

Fig. 4 displays the \( H \)-dependence of \( b \) and \( g_{\perp2} \)-values. Notice that both parameters have minimum values at the X-band field, \( H \approx 1900\) Oe.

Fig. 5 shows the X-band \( \Delta H(T) \) for \( 4.2 \leq T \leq 21\) K and \( H_{\perp} \). The data were fitted to \( \Delta H(T) = a + bT + c\delta/\exp(\delta/T) - 1 \) taking into consideration all the contributions to \( \Delta H \) in a metallic host. The 1st and 2nd terms are the same as above. The 3rd is the relaxation, also via an exchange interaction with the ce, of a thermally populated Yb\(^{3+}\) excited crystal field state at \( \delta K \) above the ground state [23]. The fitting parameters are in the inset of Fig. 5. This analysis does not consider any direct Yb\(^{3+}\) spin–phonon contribution [23]. The S and Q-band \( \Delta H(T) \) data for \( 7 \leq T \leq 20\) K also show exponential behaviors with \( c \approx 200(70) \) Oe/K and \( \delta \approx 75(20)\) K.

Within a molecular field approximation the effective \( g(T) \) may be written as \( g_{\text{eff}} = g(1 + \lambda \chi_{\perp,0}(T)) \). Fig. 6 presents a plot of \( \lambda \) vs. \( H_{\perp}(\theta) = g(\theta)/g(15) \) for \( T \lesssim 15\) K and X-band for our \( x = 0 \) crystals [15] and that from Refs. [7,24]. A linear correlation is obtained with \( \lambda \) values in the interval of \(-2\) kOe/\( \mu_B \) > \( \lambda > -3\) kOe/\( \mu_B \). In the Appendix, it is shown that the shift that gives the temperature dependence of \( g_{\text{eff}} \) arises from anisotropic exchange interactions between the Yb\(^{3+}\) ions. Fig. 7 shows the comparison between theory and experiment. Therefore, these results definitely indicate that the \( T \)-dependence of the effective \( g(x,T) \) is nothing but a consequence of the shift of the
The Sef reference the ESR of Yb3+ phase. Always observable, unless extreme bottleneck regime is achieved. In strong impurity effects, these between the Kondo ions and the /C25 H/C30. The divergence [7]. Moreover, the expected s-shift caused by the exchange interaction between the Yb3+ and ce local moments, Jce, can be estimated from the largest Korrtinga-rate value measured at Q-band in our In-flux crystal, b \cong 45 \text{ Oe/K}. Within a single band approximation [23] and absence of q-dependence of the Yb3+–ce exchange interaction, Jce(\Omega) \equiv Jce(\Omega) [26] one can write (\Delta g/\Omega^2 = \mu_b b/\pi g_k q_k), which gives: |\Delta g/\Omega| \cong 2%\). This values is far much smaller than that estimated in Ref. [7] using as a reference the ESP of Yb in the insulator PbMoO4. On the other hand, from the Korrtinga relation [23] using b \cong 45 \text{ Oe/K} and assuming a maximum bare density of state per one spin direction at the Fermi level, \eta_f, given by the Sommerfeld coefficient of the specific heat measurements (\gamma \cong 900 \text{ mJ/mol K}^2) [27] we extract a lower limit for \mu_s \cong 3 \text{ mJ}, which is about two order of magnitude larger than the value found for the Yb3+–Yb3+ exchange interaction, Jce, inferred from the molecular field parameters \theta_c and \theta_c(T) in a nearest-neighbor approximation (see Appendix) [25].

Our experiments confirm the ESP results of Sichelschmidt et al., in YbRh2Si2 below T_K \cong 25 \text{K} [7]. However, our results suggest that this ESP corresponds to a strong exchange coupled Yb3+–ce resonant collective mode. The features of this resonant collective mode resembles the bottleneck/dynamic scenario for diluted magnetic moments exchange coupled to the ce, both with g \cong 2, where in a normal metal the SLR-rate, and g-value depend on the competition between the Korrtinga/Overhauser relaxation and the ce SLR [18,29,19,28]. Then, the increase of b by the addition of non-magnetic impurities to YbRh2Si2 (Lu, Zn in Fig. 1 and La in Ref. [12]), may be associated to “opening” the bottleneck regime due to the increase in the ce spin-flip scattering [19,28]. The results and discussion about the non-magnetic Lu impurities effects and bottleneck behavior will be the subject of a forthcoming publication.

Another striking result reported in Fig. 4 is the non-monotonic H-dependence of b and effective g-value of the Yb3+–ce resonant collective mode. Admixtures via Van Vleck terms [30] may be disregarded because this contribution should scale with H. Therefore, we believe that the low H-tunability [1–3] of the ESP parameters in YbRh2Si2 is an “intrinsic” property of the NFL state near a QCP, where the strength of the Yb3+–ce magnetic coupling may be subtly tuned and allows the formation of the resonant collective mode. We attribute the absence of low H-dependent ESP results in previous reports [7] to the presence of “extrinsic” impurities and/or Rh/Si defects [15,31] that increase the SLR, b, and residual linewidth, a. Thus, as for our Zn-flux crystals, hiding the low H-dependence of the ESP parameters in the NFL phase.

In the bottleneck scenario, the Yb3+–ce resonant collective mode presents the strongest bottleneck regime (smallest b) at H \cong 1900 \text{Oe}. However, due to the subtle details of the coupling between the Kondo ions and the ce in a Kondo lattice and to strong impurity effects, these resonant collective modes may not be always observable, unless extreme bottleneck regime is achieved. The proximity to a QCP and/or the presence of enhanced spin susceptibility may favor this condition [13,32]. The bottleneck scenario for the Yb3+–ce resonant collective mode may also explain the absence of Yb3+ hyperfine ESP structure [33].

Recent calculations by Abrahams and Wolflc have suggested that the ESP linewidth may be strongly reduced by a factor involving the heavy fermion mass and quasiparticle ferromagnetic (FM) exchange interactions (m/m* = 1 – U \text{FM}_{B,P}(\Omega)) [34]. These results indicate that the estimation of the linewidth from the Kondo temperature, T_K (\Delta H = k_B T_K/\theta_k B) is an over estimation. However, these calculations may not be contemplating all the possibilities and have to be taken with care when applied to the dynamic of the ESP of YbRh2Si2 compound because, (i) it presents an AF Yb3+–Yb3+ exchange interaction (although other works in literatures have claimed in favor of the existence of FM fluctuation in YbRh2Si2 [35,36] and (ii) samples with the same thermodynamic properties present quite different linewidths (see Fig. 1). Furthermore, the anisotropy in the ESP in YbRh2Si2 reflects both single-ion crystal field effects and the Yb3+–Yb3+ and Yb3+–ce interactions.

In principle, the analysis of crystal field effects is straightforward although somewhat hindered by the inability to detect a signal when the field is along the c-axis. The anisotropy of the Yb3+–Yb3+ and Yb3+–ce is more difficult to determine and in the latter case more critical. The application of the resonant collective mode model is based on the assumption that the Yb3+–ce coupling is dominated by a scalar interaction between the Yb3+ ground state doublet pseudo-spins S_m, and the spins of the conduction electrons, s, with the consequence that the total spin s + S_m is (approximately) a constant of the motion [37,38]. In the presence of uniaxial anisotropy, only the component of the total spin along the symmetry axis is a constant of the motion. How the lower symmetry affects the formation of the collective mode is an unsolved problem requiring further study [34].

Finally, we hope that our results motivate new theoretical approaches to understand the dynamics of strong exchange coupled magnetic moments of unlike spins and g-values, as Yb3+ and ce, and explore the general existence of a resonant collective mode with a bottleneck/dynamic-like behavior.

### 4. Summary

In summary, this work reports low H-dependent ESP, below T_K \cong 25 \text{ K}, in the NFL phase of YbRh2Si2 (T \leq 10 \text{ K}). It is suggested that the observed ESP in YbRh2Si2 corresponds to a Yb3+–ce resonant collective mode in a strong bottleneck-like regime, which is highly affected by the presence of impurities and/or defects. The analysis of our data allowed us to give estimations for the Yb3+–Yb3+ exchange parameter, Jce, and a lower limit for the Yb3+–ce exchange parameter, Jce.
In applying this equation to the resonance in YbRh$_2$Si$_2$ it must be kept in mind that the g-factors and susceptibilities refer to the ground state doublet. We assume that the relevant doublet susceptibilities take the form $C_{1,2}/(T + \theta_{1,2})$ where $C_1$ and $C_2$ are constants and fit the experimental data for $\chi_{c}$ to the form $g_0 + C_{ic}/(T + \theta_{ic})$ in the temperature range of the resonance experiment (see inset of Fig. 7) with the result $\theta_{ic} = 1.48$ K. We chose $\theta_{ic}$ and the overall amplitude $g_0^c$ as adjustable parameters. The measured values $g_{ic}(T)$ were then fit to the function:

$$g_{ic}(T) = g_0^c \left[ 1 - \frac{(\theta_{ic} - \theta_{ic})}{T + \theta_{ic}} \right]^{1/2}$$

$$\approx g_0^c \left[ 1 - 0.5 \frac{(\theta_{ic} - \theta_{ic})}{T + \theta_{ic}} \right]$$

with the best fit given by $g_0^c = 3.66$ and $\theta_{ic} = 1.09$ K (see Fig. 7). The result shown in Fig. 6 is in accord with Eq. (4) and Fig. 7 since the $T$-dependent part of $\chi_{c}(T)$ is proportional to $(T + \theta_{ic})^{-1}$, the same factor that is present in Eq. (4).

The results outlined in the preceding paragraph show that the $T$-dependence of $g_{ic}(T)$ is associated with the difference in the longitudinal and transverse exchange interaction which is reflected in the difference between $\theta_{ic}$ and $\theta_{ic}$. Although it has not been possible to observe the resonance with the static field along the $c$-axis, there is a corresponding shift there as well, which takes the form:

$$g_{ic}(T) = g_0^c \left[ 1 + \frac{(\theta_{ic} - \theta_{ic})}{T + \theta_{ic}} \right]$$

Note that the shift in $g_{ic}(T)$ is in the opposite direction from the shift in $g_{ic}(T)$.

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