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Field-dependent collective ESR mode in YbRh₂Si₂

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ABSTRACT

Electron spin resonance (ESR) experiments in YbRh₂Si₂ Kondo lattice ($T_K \simeq 25$ K) at different field/frequencies ($4.1 \leq \nu \leq 34.4$ GHz) and $H_{\perp c}$ revealed: (i) a strong field dependent Yb³⁺ 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 ($\leq 15\%$) dependence of the ESR data. These results suggest that the ESR signal in YbRh₂Si₂ may be due to a coupled Yb³⁺-conduction electron resonant collective mode with a subtle field-dependent spins dynamic.

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1. Introduction

The heavy-fermion (HF) Kondo lattice YbRh₂Si₂ ($T_K \simeq 25$ K) is an antiferromagnetic (AF, $T_N = 70$ mK) tetragonal (I4/mmm) inter-metallic compound. At low- T ($T \lesssim T_K$) the magnetic susceptibility exhibits a HF behavior and at high- T ($T \gtrsim 200$ K) an anisotropic Curie–Weiss with a full Yb³⁺ magnetic moment ($\mu_{\text{eff}} \simeq 4.5 \mu_B$) is observed [1–3]. The AF ordering of YbRh₂Si₂ may be driven to $T_N \approx 0$ by fields of $H_{\perp c} \sim 650$ Oe and $H_{\parallel c} \sim 7$ 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], YbRh₂Si₂ is particularly an interesting system to study quantum criticality and NFL behavior.

Electron spin resonance (ESR) experiments at low- T ($T \lesssim 20$ K) in YbRh₂Si₂ 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, ΔH , and a g -value anisotropy consistent with Yb³⁺ in a metallic host of tetragonal symmetry. As in the early work of Tien et al. [8] the observation of a narrow Yb³⁺ 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 Yb³⁺ in stoichiometric YbRh₂Si₂ [9],

YbIr₂Si₂ [10], and YbRh₂Si₂ doped with non-magnetic impurities as Ge [11] and La [12]. Also, the ESR of Ce³⁺ in dense Kondo systems was communicated [13]. However, it is not clear yet what mechanism allows the observation of the Yb³⁺ 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 YbRh₂Si₂ ($4.2 \lesssim T \lesssim 10$ K; $0 < H \lesssim 10$ kOe). We found an unexpected H -dependence behavior of the Yb³⁺ ESR data in YbRh₂Si₂ 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_xRh₂Si₂ ($0 \leq x \leq 1.00$) 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 $\lesssim 0.015^\circ$. The electrical residual resistivity ratio, $\rho_{300\text{K}}/\rho_{1.9\text{K}}$, for the In and Zn-flux grown crystals were 35 and 10, respectively [14–16]. For the ESR spectra $\sim 2 \times 2 \times 0.5$ mm³ 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

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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 YbRh_2Si_2 ESR X-band spectra at 4.2 K and $H_{\perp c}$ for single crystals grown in In and Zn-fluxes. From the anisotropy of the field for resonance, $H_r(\theta)$ (not shown), one can obtain the angular-dependence of the effective g -value which is given by $h\nu/\mu_B H_r(\theta) = g(\theta) = [g_{\perp c}^2 \cos^2 \theta + g_{\parallel c}^2 \sin^2 \theta]^{1/2}$. From the fitting of the experimental $H_r(\theta)$ we obtain $g_{\parallel c} \lesssim 0.6(4)$ and $g_{\perp c} = 3.60(7)$. The inset of Fig. 1 displays, for $H_{\perp c}$, the Korringa-like [18,19] linear thermal broadening of the linewidth, $\Delta H(T) = a + bT$, 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 $\text{Yb}_{1-x}\text{Lu}_x\text{Rh}_2\text{Si}_2$ at 4.2 K and $H_{\perp c}$ 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 $x \leq 0.15$ the Yb^{3+} ESR intensity is nearly constant, for $0.15 < x \leq 1.00$ the intensity vanish completely and no ESR could be detected. It is worth mention that for $x > 0.15$ and $T \gtrsim 200$ K, $\chi_{\perp c}(T)$ follows a Curie–Weiss law with a full Yb^{3+} magnetic moment and that for $x \leq 0.15$ 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 YbRh_2Si_2 ,

Fig. 2. X-band Yb^{3+} relative normalized integrated ESR intensities, $I_{4.2}(x)/I_{4.2}(0)$, at 4.2 K and $H_{\perp c}$ for $\text{Yb}_{1-x}\text{Lu}_x\text{Rh}_2\text{Si}_2$.

Fig. 1. (Color online) ESR spectra at 4.2 K and $H_{\perp c}$ for single crystals grown in In and Zn-fluxes and for the In-flux crystals of Ref. [7]. Inset: $\Delta H(T) = a + bT$ for Zn-flux crystals and $H_{\perp c}$ at X and Q-bands.

Fig. 3. (Color online) X, S and Q-bands low- T dependence of the Yb^{3+} ESR for $H_{\perp c}$: (a) $\Delta H(T)$ and (b) effective $g(T)$ -value. The lines represent the linear fit in (a) and a guide to the eyes in (b).

measured at S, X and Q-bands for $H_{\perp c}$. 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 $g_{4,2}$ -values also at the X-band. The effective g -value accuracy is much higher than that obtained from $h\nu/\mu_B H_r(\theta) = g(\theta) = [g_{\perp c}^2 \cos^2 \theta + g_{\parallel c}^2 \sin^2 \theta]^{1/2}$ because proper experimental conditions were chosen for these $H_{\perp c}$ measurements.

Fig. 4 displays the H -dependence of b and $g_{4,2}$ -values. Notice that both parameters have minimum values at the X-band field, $H \approx 1900$ Oe.

Fig. 4. (Color online) Yb^{3+} ESR H -dependence (X, S and Q-bands) of b and effective $g_{4,2}$ -value for $H_{\perp c}$.

Fig. 5. (Color online) Yb^{3+} ESR X-band $\Delta H(T)$ for $H_{\perp c}$ and $4.2 \leq T \leq 21$ K. The fitting parameters correspond to the best fit to $\Delta H(T) = a + bT + c\delta/[\exp(\delta/T) - 1]$.

Fig. 5 shows the X-band $\Delta H(T)$ for $4.2 \leq T \leq 21$ K and $H_{\perp c}$. The data were fitted to $\Delta H(T) = a + bT + c\delta/[\exp(\delta/T) - 1]$ taking into consideration all the contributions to ΔH 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 δ 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 $g_{\text{eff}} = g(1 + \lambda\chi_{\perp c}(T))$. Fig. 6 presents a plot of $\Delta g/g = (g_{\text{eff}}(T) - g(15))/g(15) \propto \lambda\chi_{\perp c}(T)$ for $T \leq 15$ K and X-band for our $x = 0$ crystals [15] and that from Refs. [7,24]. A linear correlation is obtained with λ values in the interval of $-2 \text{ kOe}/\mu_B > \lambda > -3 \text{ kOe}/\mu_B$. In the Appendix, it is shown that the shift that gives the temperature dependence of g_{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

Fig. 6. (Color online) effective g -shift and magnetic susceptibility correlation, $(g_{\text{eff}} - g)/g \propto \lambda\chi_{\perp c}(T)$, for $T \leq 15$ K and X-band for $x = 0$ crystals (see text).

Fig. 7. $g_{\perp c}(T)$ vs. T . The solid line is the theoretical curve obtained from Eq. (4). The inset shows the fit of $\chi_{\perp c}^1(T)$ to a Curie–Weiss law.

field for resonance toward higher fields due to an AF internal molecular field and has nothing to do with a $\Delta g = c/\ln(T_K/T)$ divergence [7]. Moreover, the expected g -shift caused by the exchange interaction between the Yb^{3+} and ce local moments, J_{fce} , can be estimated from the largest Korringa-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 Yb^{3+} – ce exchange interaction, $J_{fce}(\mathbf{q}) \cong J_{fce}(\mathbf{0})$, [26] one can write $(\Delta g/g)^2 = \mu_B b / \pi g k_B$, which gives $|\Delta g/g| \lesssim 2\%$. This value is far much smaller than that estimated in Ref. [7] using as a reference the ESR of Yb^{3+} in the insulator PbMoO_4 . On the other hand, from the Korringa 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, η_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 $|J_{fce}| \gtrsim 3 \text{ meV}$, which is about two orders of magnitude larger than the value found for the Yb^{3+} – Yb^{3+} exchange interaction, $|J_{ff}|$, inferred from the molecular field parameters θ_{lc} and $\theta_{\perp c}(T)$ in a nearest-neighbor approximation (see Appendix) [25].

Our experiments confirm the ESR results of Sichelschmidt et al., in YbRh_2Si_2 below $T_K \cong 25 \text{ K}$ [7]. However, our results suggest that this ESR corresponds to a strong exchange coupled Yb^{3+} – 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, b , and g -value depend on the competition between the Korringa/Overhauser relaxation and the ce SLR [18,29,19,28]. Then, the increase of b by the addition of non-magnetic impurities to YbRh_2Si_2 (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 Yb^{3+} – 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 ESR parameters in YbRh_2Si_2 is an “intrinsic” property of the NFL state near a QCP, where the strength of the Yb^{3+} – ce magnetic coupling may be subtly tuned and allows the formation of the resonant collective mode. We attribute the absence of low H -dependent ESR 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 ESR parameters in the NFL phase.

In the bottleneck scenario, the Yb^{3+} – ce resonant collective mode presents the strongest bottleneck regime (smallest b) at $H \approx 1900 \text{ Oe}$. 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 Yb^{3+} – ce resonant collective mode may also explain the absence of Yb^{3+} hyperfine ESR structure [33].

Recent calculations by Abrahams and Wolfle have suggested that the ESR linewidth may be strongly reduced by a factor involving the heavy fermion mass and quasiparticle ferromagnetic (FM) exchange interactions $(m/m^*)(1 - U\chi_{ff,H}^+(0))$ [34]. These results indicate that the estimation of the linewidth from the Kondo temperature, T_K ($\Delta H = k_B T_K / g\mu_B$) 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 ESR of YbRh_2Si_2 compound because, (i) it presents an AF Yb^{3+} – Yb^{3+} exchange interaction (although other works in literatures have claimed in favor of the existence of FM fluctuation in YbRh_2Si_2 [35,36] and (ii) samples with the same thermodynamic properties present quite different linewidths (see Fig. 1). Furthermore, the anisotropy in the ESR in YbRh_2Si_2 reflects both single-ion crystal field effects and the Yb^{3+} – Yb^{3+} and Yb^{3+} – 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 Yb^{3+} – Yb^{3+} and Yb^{3+} – 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 Yb^{3+} – ce coupling is dominated by a scalar interaction between the Yb^{3+} ground state doublet pseudo-spins S_{ps} , and the spins of the conduction electrons, s , with the consequence that the total spin $s + S_{ps}$ 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 Yb^{3+} 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 ESR, below $T_K \cong 25 \text{ K}$, in the NFL phase of YbRh_2Si_2 ($T \lesssim 10 \text{ K}$). It is suggested that the observed ESR in YbRh_2Si_2 corresponds to a Yb^{3+} – 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 Yb^{3+} – Yb^{3+} exchange parameter, J_{ff} , and a lower limit for the Yb^{3+} – ce exchange parameter, $|J_{fce}|$.

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Appendix

In Fig. 6, it was noted that g_{eff} depends linearly on the transverse susceptibility. In this appendix we show the origin of the linear dependence. The starting point is a general theory for magnetic resonance in anisotropic magnets [39] which leads to an expression for the effective g -factor in an orthorhombic crystal that can be written in the form:

$$g_{\text{eff}} = \frac{g_b^M g_c^M \chi_a}{g_a^M (\chi_a \chi_b)^{1/2}} \quad (1)$$

when the static field is along the a -axis. Here the microscopic g -factors are denoted by $g_{a,b,c}^M$ and the corresponding static susceptibilities by $\chi_{a,b,c}$. In the case of a uniaxial system with the static field perpendicular to the c -axis, (1) reduces to

$$g_{\text{eff}\perp c} = g_c^M \left[\frac{\chi_{\perp c}}{\chi_{\parallel c}} \right]^{1/2} \quad (2)$$

In applying this equation to the resonance in YbRh_2Si_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 $C_{\perp,\parallel}/(T + \theta_{\perp,\parallel})$ where $C_{\perp}$ and $C_{\parallel}$ are constants and fit the experimental data for $\chi_{\perp,c}$ to the form $\chi_0 + C_{\perp,c}/(T + \theta_{\perp,c})$ in the temperature range of the resonance experiment (see inset of Fig. 7) with the result $\theta_{\perp,c} = 1.48$ K. We chose $\theta_{\parallel,c}$ and the overall amplitude $g_{\perp,c}^0$ as adjustable parameters. The measured values $g_{\perp,c}(T)$ were then fit to the function:

$$g_{\perp,c}(T) = g_{\perp,c}^0 \left[1 - \left(\frac{\theta_{\perp,c} - \theta_{\parallel,c}}{T + \theta_{\perp,c}} \right) \right]^{1/2} \quad (3)$$

$$\approx g_{\perp,c}^0 \left[1 - 0.5 \left(\frac{\theta_{\perp,c} - \theta_{\parallel,c}}{T + \theta_{\perp,c}} \right) \right] \quad (4)$$

with the best fit given by $g_{\perp,c}^0 = 3.66$ and $\theta_{\parallel,c} = 1.09$ 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 $\chi_{\perp,c}(T)$ is proportional to $(T + \theta_{\perp,c})^{-1}$, the same factor that is present in Eq. (4).

The results outlined in the preceding paragraph show that the T -dependence of $g_{\perp,c}(T)$ is associated with the difference in the longitudinal and transverse exchange interaction which is reflected in the difference between $\theta_{\perp,c}$ and $\theta_{\parallel,c}$. 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:

$$g_{\parallel,c}(T) = g_{\parallel,c}^0 \left[1 + \left(\frac{\theta_{\perp,c} - \theta_{\parallel,c}}{T + \theta_{\parallel,c}} \right) \right] \quad (5)$$

Note that the shift in $g_{\parallel,c}(T)$ is in the opposite direction from the shift in $g_{\perp,c}(T)$.

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