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# Field-dependent collective ESR mode in $YbRh_2Si_2$

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ABSTRACT

Electron spin resonance (ESR) experiments in $YbRh_2Si_2$ 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 $Yb^{3+}$ 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 ($\leq 15\%$) dependence of the ESR data. These results suggest that the ESR signal in $YbRh_2Si_2$ 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 $YbRh_2Si_2$ ($T_K \simeq 25\,K$) is an antiferrromagnetic (AF, $T_N = 70\,mK$) tetragonal (I4/mmm) intermetallic 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 $Yb^{3+}$ magnetic moment ($\mu_{eff} \simeq 4.5\mu_B$) is observed [1–3]. The AF ordering of $YbRh_2Si_2$ may be driven to $T_N \approx 0$ by fields of $H_{\perp c}$~650 Oe and $H_{\parallel c}$~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], $YbRh_2Si_2$ 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 $YbRh_2Si_2$ 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 $YbRh_2Si_2$ [9], $YbIr_2Si_2$ [10], and $YbRh_2Si_2$ 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 $YbRh_2Si_2$ ($4.2 \lesssim T \lesssim 10\,K$; $0 < H \lesssim 10\,kOe$). We found an unexpected $H$-dependence behavior of the $Yb^{3+}$ ESR data in $YbRh_2Si_2$ that, we hope, will contribute to the understanding of the observed ESR signal in this system.

## 2. Experiment

Single crystals of $Yb_{1-x}Lu_xRh_2Si_2$ ($0 \leq x \lesssim 1.00$) 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, $\rho_{300\,K}/\rho_{1.9\,K}$, for the In and Zn-flux grown crystals were 35 and 10, respectively [14–16]. For the ESR spectra $\sim 2 \times 2 \times 0.5\,mm^3$ 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

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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 $Yb_{1-x}Lu_xRh_2Si_2$ 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 $0.15 < x \lesssim 1.00$ 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 an 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 $\Delta H$ and *effective* g-value of the $Yb^{3+}$ ESR in In-flux grown $YbRh_2Si_2$,

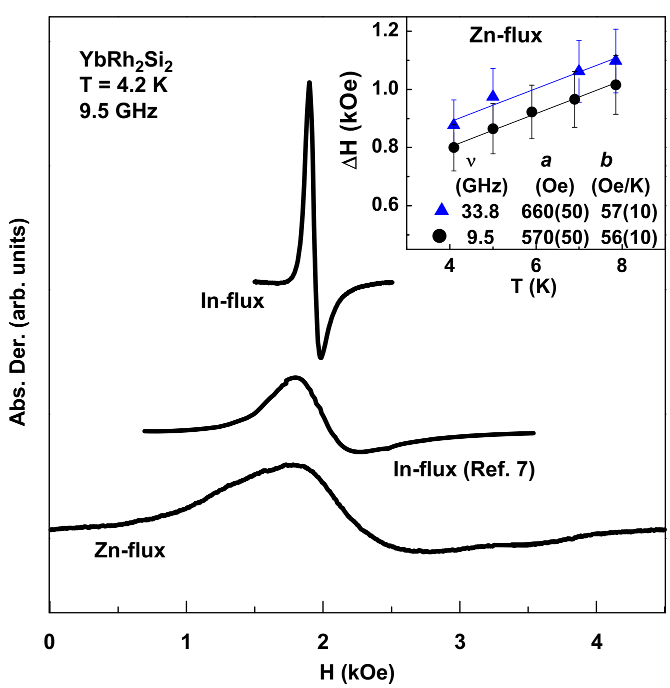


**Fig. 1.** (Color online) ESR spectra at 4.2 K and $H_{\perp c}$ for single crystals grown in In and Zn-fluxes and for the In-flux crystals of Ref. [7]. Inset: $\Delta H(T) = a + bT$ for Zn-flux crystals and $H_{\perp c}$ at X and Q-bands.

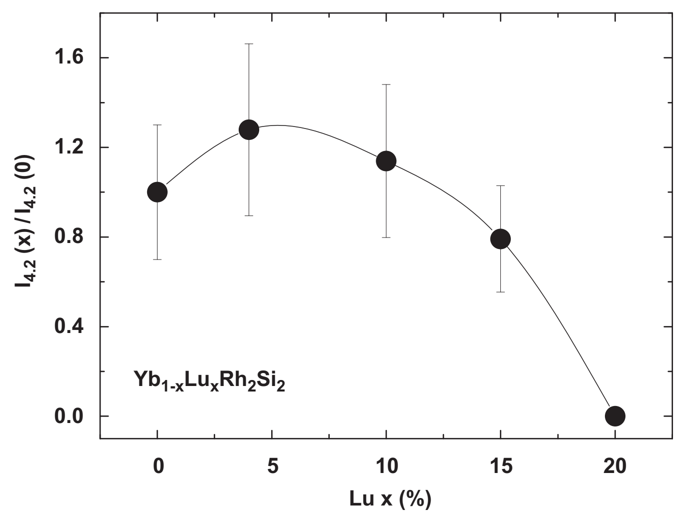


**Fig. 2.** X-band $Yb^{3+}$ relative normalized integrated ESR intensities, $I_{4.2}(x)/I_{4.2}(0)$, at 4.2 K and $H_{\perp c}$ for $Yb_{1-x}Lu_xRh_2Si_2$.

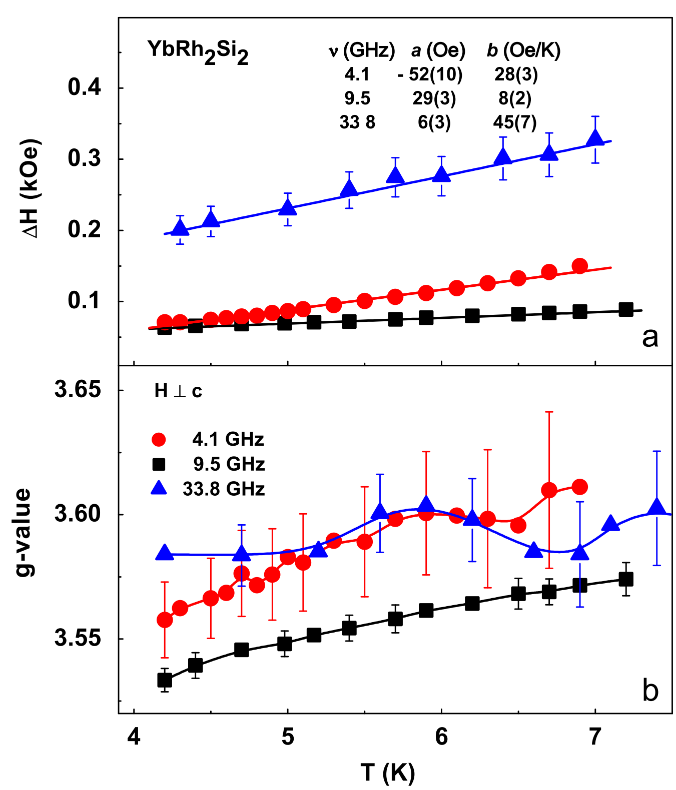


**Fig. 3.** (Color online) X, S and Q-bands low-$T$ dependence of the $Yb^{3+}$ ESR for $H_{\perp c}$: (a) $\Delta H(T)$ and (b) *effective* g($T$)-value. The lines represent the linear fit in (a) and a guide to the eyes in (b).

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, $H \simeq 1900$ 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 consider 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 $h\nu/\mu_B H_r(\theta) = g(\theta) = [g^2_{\perp c}\cos^2\theta + g^2_{\parallel c}\sin^2\theta]^{1/2}$ 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, $H \simeq 1900$ Oe.

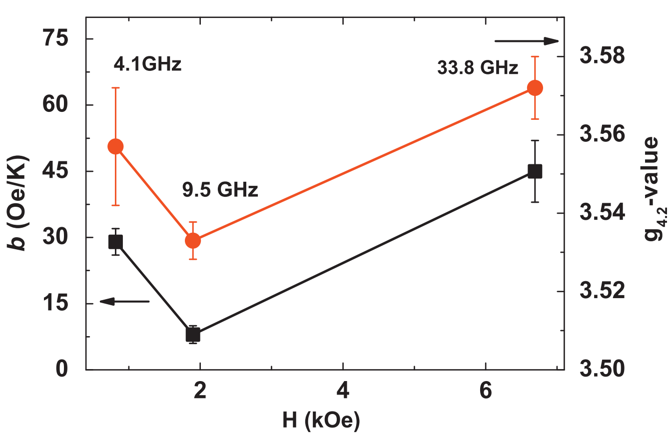


**Fig. 4.** (Color online) $Yb^{3+}$ ESR $H$-dependence (X, S and Q-bands) of $b$ and *effective* $g_{4.2}$-value for $H_{\perp c}$.

Fig. 5 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 $\Delta H$ 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 $7 \lesssim T \lesssim 20$ 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 $g_{eff} = g(1 + \lambda\chi_{\perp c}(T))$. Fig. 6 presents a plot of $\Delta g/g = (g_{eff}(T) - g(15))/g(15) \propto \lambda\chi_{\perp c}(T)$ 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 $\lambda$ values in the interval of $-2\,\mathrm{kOe}/\mu_B > \lambda > -3\,\mathrm{kOe}/\mu_B$. 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

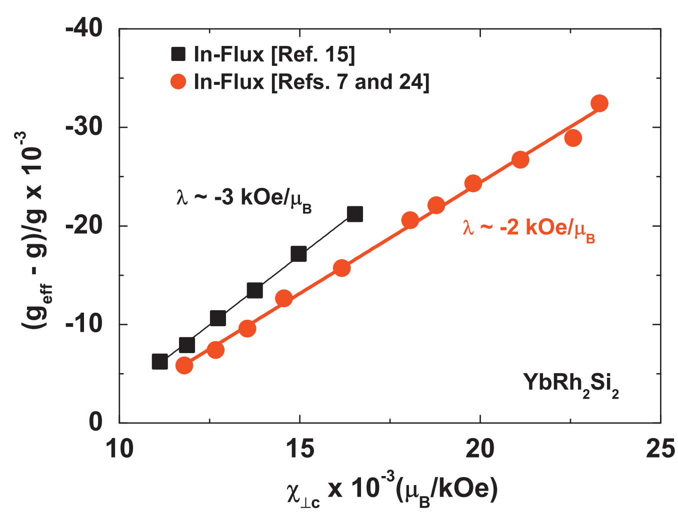


**Fig. 6.** (Color online) *effective* $g$-shift and magnetic susceptibility correlation, $(g_{eff} - g)/g \propto \lambda\chi_{\perp c}(T)$, for $T \leq 15$ K and X-band for $x = 0$ crystals (see text).

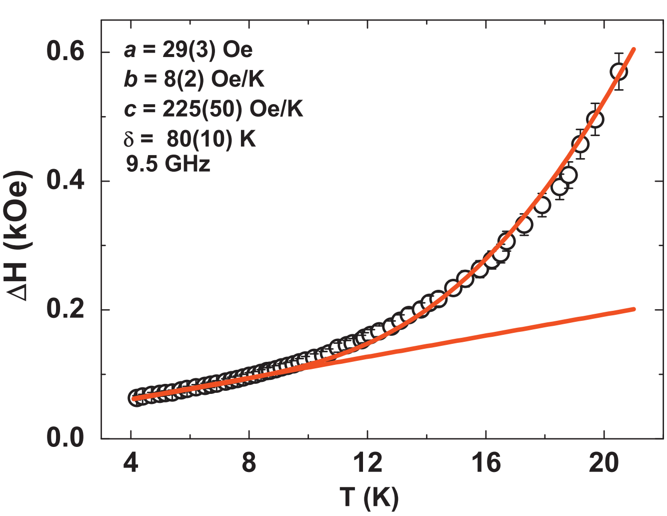


**Fig. 5.** (Color online) $Yb^{3+}$ ESR X-band $\Delta H(T)$ for $H_{\perp c}$ and $4.2 \leq T \leq 21$ K. The fitting parameters correspond to the best fit to $\Delta H(T) = a + bT + c\delta/[\exp(\delta/T) - 1]$.

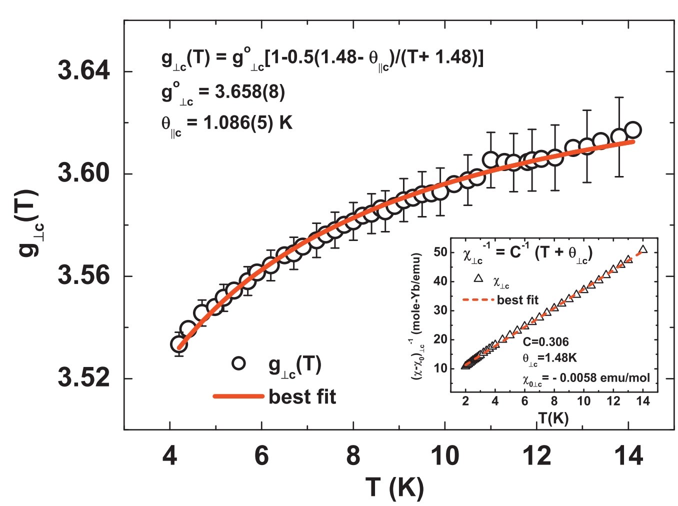


**Fig. 7.** $g_{\perp c}(T)$ vs. $T$. The solid line is the theoretical curve obtained from Eq. (4). The inset shows the fit of $\chi^{-1}_{\perp c}(T)$ to a Curie–Weiss law.

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, $b \cong 45\,\mathrm{Oe/K}$. 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 values 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 $b \cong 45\,\mathrm{Oe/K}$ and assuming a maximum *bare* density of state per one spin direction at the Fermi level, $\eta_F$, given by the Somerfield coefficient of the specific heat measurements ($\gamma \cong 900\,\mathrm{mJ/mol\,K^2}$) [27] we extract a lower limit for $|J_{fce}| \gtrsim 3\,\mathrm{meV}$, which is about two order of magnitude larger than the value found for the $Yb^{3+}$–$Yb^{3+}$ exchange interaction, $|J_{ff}|$, inferred from the molecular field parameters $\theta_{\parallel c}$ 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 $T_K \simeq 25\,\mathrm{K}$ [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 $H \approx 1900\,\mathrm{Oe}$. 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 $(m/m^*)(1 - \tilde{U}\chi^{+-}_{ff,H}(0))$ [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 quit 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 $T_K \simeq 25\,\mathrm{K}$, in the NFL phase of $YbRh_2Si_2$ ($T \lesssim 10\,\mathrm{K}$). 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:

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

when the static field is along the $a$-axis. Here the microscopic $g$-factors are denoted by $g^M_{a,b,c}$ 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

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

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 $\theta_{\perp c} = 1.48\,\mathrm{K}$. We chose $\theta_{\parallel c}$ and the overall amplitude $g^0_{\perp c}$ as adjustable parameters. The measured values $g_{\perp c}(T)$ were then fit to the function:

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

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

with the best fit given by $g^0_{\perp c} = 3.66$ and $\theta_{\parallel c} = 1.09\,\mathrm{K}$ (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:

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

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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