

## Magnetic field dependence and bottlenecklike behavior of the ESR spectra in $\text{YbRh}_2\text{Si}_2$

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Electron spin resonance (ESR) experiments at different fields or frequencies ( $4.1 \leq \nu \leq 34.4$  GHz) in the Kondo lattice ( $T_K \approx 25$  K)  $\text{YbRh}_2\text{Si}_2$  single-crystal compounds confirmed the observation of a single anisotropic Dysonian resonance with  $g_{\perp c} \approx 3.55$  and no hyperfine components for  $4.2 \leq T \leq 20$  K. However, our studies differently reveal that (i) the ESR spectra for  $H_{\perp c}$  show strong-field-dependent spin-lattice relaxation, (ii) a weak-field and temperature-dependent *effective g* value, (iii) a dramatic 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 a different scenario where the ESR signal may be associated to a coupled  $\text{Yb}^{3+}$ -conduction electron *resonant collective mode* with a strong bottleneck and dynamiclike behavior.

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### I. INTRODUCTION

The metallic antiferromagnetic [(AF)  $T_N = 70$  mK] tetragonal ( $I4/mmm$ ) heavy-fermion (HF) Kondo lattice ( $T_K \approx 25$  K)  $\text{YbRh}_2\text{Si}_2$  is a peculiar system that has attracted great attention of the scientific community interested in strongly correlated electron systems. At high- $T$  ( $T \geq 200$  K) its magnetic susceptibility follows an anisotropic Curie-Weiss law with a full  $\text{Yb}^{3+}$  magnetic moment ( $\mu_{\text{eff}} \approx 4.5\mu_B$ ), but at low  $T$  ( $T \leq T_K$ ) HF behavior is found.<sup>1,3,34</sup> The AF ordering<sup>2</sup> of  $\text{YbRh}_2\text{Si}_2$  can be suppressed by a weak magnetic field of  $H_{\perp c} \sim 650$  Oe and when this AF state is  $H$  tuned toward a quantum critical point (QCP), pronounced non-Fermi-liquid (NFL) behavior is found.<sup>1,34</sup> These properties made  $\text{YbRh}_2\text{Si}_2$  a special system for studying quantum criticality and NFL behavior in connection to other systems near a QCP.<sup>4,5</sup>

Other unexpected and interesting aspect of  $\text{YbRh}_2\text{Si}_2$  was revealed by electron spin resonance (ESR) studies.<sup>6</sup> 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  $\text{Yb}^{3+}$  in a metallic host of tetragonal symmetry was found for  $T \leq 20$  K. However, despite its  $H$ -tunable ground state, no  $H$ -dependent ESR parameters were reported for  $H \leq 10$  kOe.<sup>6</sup> Moreover, a narrow  $\text{Yb}^{3+}$  ESR in a dense Kondo system as  $\text{YbRh}_2\text{Si}_2$  below  $T_K$  was totally unexpected. Since its first observation<sup>6</sup> various reports were published on the ESR of  $\text{Yb}^{3+}$  in stoichiometric  $\text{YbRh}_2\text{Si}_2$ ,<sup>7</sup>  $\text{YbIr}_2\text{Si}_2$ ,<sup>8</sup> and  $\text{YbRh}_2\text{Si}_2$  doped with non-magnetic impurities as Ge (Ref. 9) and La.<sup>10</sup> Recently, the ESR of  $\text{Ce}^{3+}$  in dense Kondo systems was also communicated.<sup>11</sup> Nevertheless, there is still no clear understanding of the microscopic mechanism that allows the observation of such a narrow  $\text{Yb}^{3+}$  ESR line characteristic of a local moment in a metallic host.

In this work, our main report is on the  $H$ -dependent ESR experiments in the NFL phase of  $\text{YbRh}_2\text{Si}_2$  ( $4.2 \leq T \leq 10$  K;  $0 < H \leq 10$  kOe). Our results reveal an unconventional  $H$  dependence and bottlenecklike behavior of the  $\text{Yb}^{3+}$  resonance in this system that may help to shed light on the origin of such unexpected ESR signal.

### II. EXPERIMENT

Single crystals of  $\text{Yb}_{1-x}\text{Lu}_x\text{Rh}_2\text{Si}_2$  ( $0 \leq x \leq 1.00$ ) were grown from In and Zn fluxes as reported.<sup>12–14</sup> 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 [(0,0,4) Bragg peak] for various crystals which revealed a mosaic structure of maximum  $c$ -axis angular spread of  $\leq 0.015^\circ$  and a correlation length of  $\sim 650$  nm (grain size). 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.<sup>12–14</sup> The ESR spectra were taken in  $\sim 2 \times 2 \times 0.5$  mm<sup>3</sup> single crystals in a Bruker S, X, and Q bands (4.1, 9.5, and 33.8 GHz) spectrometer using appropriated resonators and  $T$ -controller systems.

Single resonance of a Kramers doublet ground state with no hyperfine components was observed at all bands. The Dysonian line shape ( $A/B \approx 2.5$ ) corresponds to a microwave skin depth smaller than the size of the crystals.<sup>15</sup>

### III. RESULTS AND DISCUSSION

Figure 1 presents at  $T = 4.2$  K (a) the  $\text{Yb}^{3+}$  ESR  $\theta$  dependence of the field for resonance  $H_r(\theta)$  and (b) linewidth  $\Delta H(\theta)$  in a plane perpendicular to the  $ab$  plane for the In-flux  $\text{YbRh}_2\text{Si}_2$  at S, X, and Q bands. The  $\theta$  dependence of the *effective g* value can be inferred from  $H_r(\theta)$  and it is given by  $h\nu/\mu_B H_r(\theta) = g(\theta) = [g_{\perp c}^2 \cos^2 \theta + g_{\parallel c}^2 \sin^2 \theta]^{1/2}$ . The solid

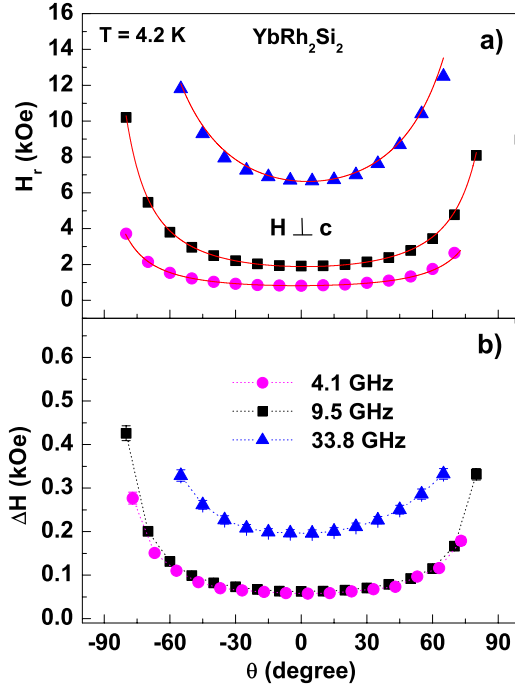


FIG. 1. (Color online) X, S, and Q-band angular dependences of the  $\text{Yb}^{3+}$  ESR at 4.2 K in a plane perpendicular to the  $ab$  plane: (a)  $H_r(\theta)$  (solid lines, see text) and (b)  $\Delta H(\theta)$  (dashed lines are guides for the eyes).

lines in Fig. 1(a) are simultaneous fittings for the three bands of the  $H_r(\theta)$  data to  $h\nu/\mu_B g(\theta)$ . These fittings give  $g_{\parallel c} \leq 0.6(4)$  and  $g_{\perp c} = 3.60(7)$ . Figure 1(b) shows that  $\Delta H(\theta)$  for the S and X bands are about the same, indicating the absence of appreciable low  $H$ -dependent inhomogeneous broadening. At Q band, in contrast,  $\Delta H(\theta)$  is significantly broader, suggesting the presence of a  $H$ -dependent broadening mechanism (see below). Furthermore, for the three bands and away from the  $ab$  plane,  $\Delta H$  broadens by  $\approx 200$  Oe. The  $g_{\parallel c}$ -value distribution due to an angle spread of  $\approx 0.015^\circ$  for the  $c$  axis (mosaic effect) would only contribute to an inhomogeneous broadening of  $\leq 5$  Oe and cannot account for the broadening of  $\approx 200$  Oe.

Figures 2(a) and 2(b) show, respectively, the low- $T$  dependences of  $\Delta H$  and effective  $g$  value of the  $\text{Yb}^{3+}$  ESR in In-flux-grown  $\text{YbRh}_2\text{Si}_2$  measured at the three bands for  $H_{\perp c}$ . In this  $T$  interval and within the error bars, it is found  $\Delta H = a + bT$  for the three bands. This suggests a Korringa type of mechanism for the  $\text{Yb}^{3+}$  spin-lattice relaxation (SLR), i.e., the  $\text{Yb}^{3+}$  local moment is exchange coupled to the conduction electrons (ces).<sup>16</sup> The residual linewidth  $a$  and relaxation rate  $b = \Delta H/\Delta T$  are given in Fig. 2(a). 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. However, the large  $\Delta H(\theta)$  measured at Q band and at  $T = 4.2$  K, relative to those in S and X bands [see Fig. 1(b)], is probably associated to a homogeneous ( $H$ -induced) increase in the SLR rate  $b$  and to a weak  $H$ -dependent inhomogeneous broadening of the residual linewidth. A  $H$ -dependent SLR rate  $b$  is not expected for a normal local magnetic

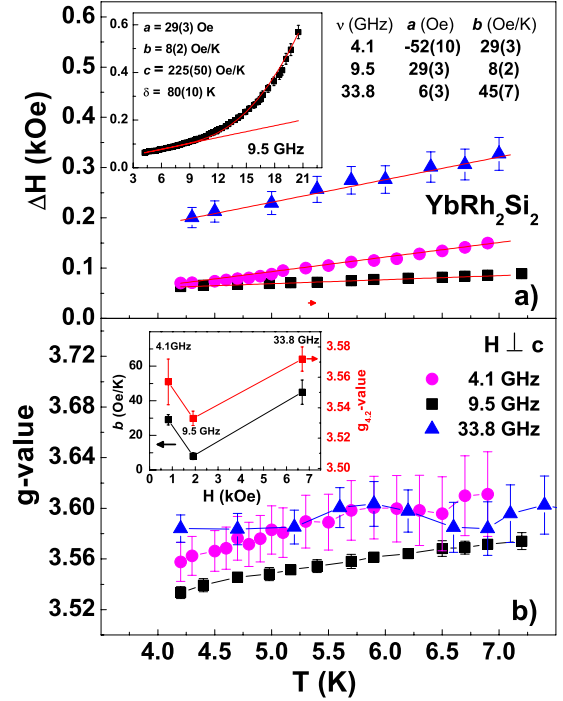


FIG. 2. (Color online) X, S, and Q-band low- $T$  dependences of the  $\text{Yb}^{3+}$  ESR for  $H_{\perp c}$  (a)  $\Delta H(T)$  and (b) effective  $g(T)$  value. Inset of (a): X-band  $\Delta H(T)$  for  $4.2 \leq T \leq 21$  K and fitting parameters to  $\Delta H(T) = a + bT + c\delta/[\exp(\delta/T) - 1]$ . Inset of (b):  $H$  dependence of  $b$  and effective  $g_{4.2}$  value.

moment-ce exchange-coupled system, where the Korringa rate is frequency and field independent.<sup>17</sup> However, since the  $\text{Yb}^{3+}$  and ce magnetic moments carry unlike spins and  $g$  values, the  $H$  dependence of  $b$ , having a minimum value at the X-band field  $H \approx 1900$  Oe [see inset of Fig. 2(b)], may be an anomalous manifestation of a bottlenecklike behavior. Figure 2(b) shows that the  $T$  dependence of the effective  $g$  values is 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  $g(\theta)$  of Fig. 1 because proper experimental conditions were chosen for these  $H_{\perp c}$  measurements. The inset of Fig. 2(a) shows the X-band  $\Delta H(T)$  for  $4.2 \leq T \leq 21$  K. 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 first and second terms are the same as above. The third is the relaxation, also via an exchange interaction with the (ce), of a thermally populated  $\text{Yb}^{3+}$  excited crystal-field state at  $\delta$  K above the ground state.<sup>18</sup> The fitting parameters are in the inset of Fig. 2(a). This analysis does not consider any direct  $\text{Yb}^{3+}$  spin-phonon contribution.<sup>18</sup> 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.

Figure 3 shows the X-band low- $T$   $\text{Yb}^{3+}$  ESR in  $\text{Yb}_{1-x}\text{Lu}_x\text{Rh}_2\text{Si}_2$ : (a)  $\Delta H(T)$  for  $x=0$  at different  $\theta$  between  $H_{\perp c}$  and  $H_{\parallel c}$ , (b)  $\Delta H(T)$  for  $0 \leq x \leq 0.15$  and  $H_{\perp c}$ , and (c) the effective  $g(x, T)$  for  $0 \leq x \leq 0.10$ .  $\Delta H(T)$  was fitted to  $\Delta H(T) = a + bT$  with parameters given in the insets of Figs. 3(a), 3(b), and 5(a). For  $7 \text{ K} \leq T \leq 20 \text{ K}$ ,  $\Delta H(T)$  also shows exponential behaviors with parameters  $c \approx 200(80)$  Oe/K and

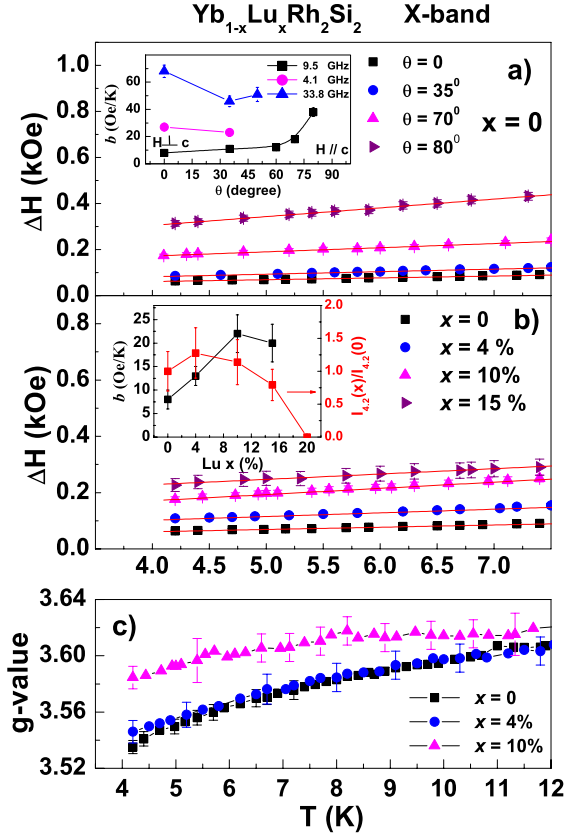


FIG. 3. (Color online) X-band low- $T$   $\text{Yb}^{3+}$  ESR data for  $\text{Yb}_{1-x}\text{Lu}_x\text{Rh}_2\text{Si}_2$ : (a)  $\Delta H(T)$  for  $\theta$  between  $H_{\perp c}$  and  $H_{\parallel c}$  and  $x=0$ , (b)  $\Delta H(T,x)$ , and (c) effective  $g(x,T)$  for various Lu concentrations and  $H_{\perp c}$ . The parameters of  $\Delta H(T)=a+bT$  are in the insets of (a) and (b) and in Fig. 5(a). Inset of (a):  $b(\theta)$  for the  $S$  and  $Q$  bands. Inset of (b): relative normalized ESR integrated intensities at 4.2 K,  $I_{4.2}(x)/I_{4.2}(0)$ .

$\delta \approx 65(30)$  K.<sup>18</sup> The inset of Fig. 3(a) includes  $b(\theta)$  for the  $S$  and  $Q$  bands and inset of Fig. 3(b) presents the relative normalized X-band ESR integrated intensities at 4.2 K 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. The X-band data show that  $b$  increases as  $H$  gets close to  $H_{\parallel c}$  and as  $x$  increases.  $g(x,T)$  and  $g(0,T)$  are similar but with higher values as  $x$  increases. Again, the  $\theta$  and  $x$  dependences of  $b$  and  $T$  dependences of the effective  $g$  values resemble a bottleneck and/or dynamiclike behavior.<sup>19</sup> Furthermore, the inset of Fig. 3(b) shows that while for  $x \leq 0.15$  there is nearly no decrease in the  $\text{Yb}^{3+}$  ESR intensity, for  $0.15 < x \leq 1.00$  the intensity drops dramatically and no ESR could be detected; although, interestingly, for  $x > 0.15$  and  $T \geq 200$  K,  $\chi_{\perp c}(T)$  follows a Curie-Weiss law with a full  $\text{Yb}^{3+}$  magnetic moment. We should mention that for  $x \leq 0.15$  there is no appreciable changes in the thermodynamic properties of these compounds.<sup>12,13</sup> The absence of resonance for  $x > 0.15$  strongly suggests that the observed ESR for  $x < 0.15$  cannot be associated to a single  $\text{Yb}^{3+}$  ion resonance but rather to a *resonant collective mode* of exchange-coupled  $\text{Yb}^{3+}$ -ce magnetic moments. We argue that a strong  $\text{Yb}^{3+}$ -ce exchange coupling may broaden and shift the ce resonance toward the

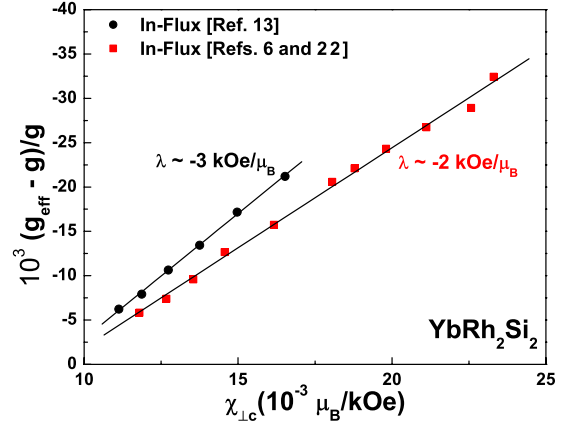


FIG. 4. (Color online) Effective  $g$  shift and magnetic-susceptibility correlation,  $(g_{\text{eff}} - g)/g \propto \lambda \chi_{\perp c}(T)$ , for  $T \leq 15$  K and X band for  $x=0$  crystals (see text).

$\text{Yb}^{3+}$  resonance, building up a  $\text{Yb}^{3+}$ -ce coupled mode with bottleneck and/or dynamiclike features. An internal field caused by the  $\text{Yb}^{3+}$  local moments may be responsible for the shift of the ce resonance.<sup>20</sup> Moreover, the Lu doping may disrupt the collective-mode coherence and open the bottleneck and/or dynamic regime.<sup>19</sup>

Moreover, within a molecular-field approximation the effective  $g(x,T)$  may be written as  $g_{\text{eff}} = g[1 + \lambda \chi_{\perp c}(T)]$ . Figure 4 presents a plot of  $\Delta g/g = (g_{\text{eff}} - g)/g \propto \lambda \chi_{\perp c}(T)$  for  $T \leq 15$  K and X band for our  $x=0$  crystals<sup>13</sup> and that from Refs. 6 and 22. A linear correlation is obtained with  $\lambda$  values in the interval  $-2 \text{ kOe}/\mu_B > \lambda > -3 \text{ kOe}/\mu_B$  which corresponds to a  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  exchange interaction of  $J_{ff} \sim -200$  mK ( $\sim -0.02$  meV) within a first-neighbor mean-field approximation.<sup>21</sup> A Néel temperature of  $T_N \equiv J_{ff}(g/2)^2 \sim 500$  mK can be roughly estimated from these values. 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 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.<sup>6</sup> Moreover, the expected  $g$  shift caused by the exchange interaction between the  $\text{Yb}^{3+}$  and ce local moments  $J_{fce}$  can be estimated from the largest measured Korringa-rate value (unbottleneck),  $b \approx 40$  Oe/K. Within a single band approximation<sup>18</sup> and absence of  $q$  dependence of the  $\text{Yb}^{3+}$ -ce exchange interaction  $J_{fce}(\mathbf{q}) \equiv J_{fce}(\mathbf{0})$ ,<sup>23</sup> one can write  $(\Delta g/g)^2 = \mu_B b / \pi g k_B$ , which gives  $|\Delta g/g| \approx 2\%$ . This value is far much smaller than that estimated in Ref. 6 using as a reference the ESR of  $\text{Yb}^{3+}$  in the insulator  $\text{PbMoO}_4$ . On the other hand, from the Korringa relation<sup>18</sup> using  $b \approx 40$  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 ( $\gamma \approx 900$  mJ/mol K<sup>2</sup>) (Ref. 24), we extract a lower limit for  $|J_{fce}| \geq 3$  meV, which is about 2 orders of magnitude larger than the value found for the  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  exchange interaction  $|J_{ff}|$  (see above).

The behavior of the effective  $g(x,T)$  shown in Fig. 3(c) is also qualitatively consistent with a collective-mode interpretation in that the effective  $g$  factors lie between the  $\text{Yb}^{3+}$  value of  $g \approx 3.6$  and the ce value of  $g \approx 2$ . The effective  $g$

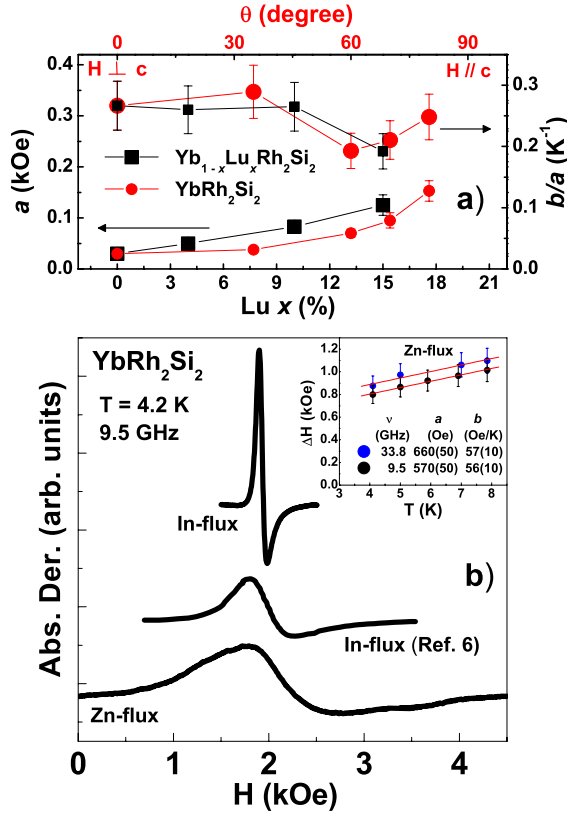


FIG. 5. (Color online) (a) X-band  $Yb^{3+}$  ESR:  $a(x)$ ,  $a(\theta)$ ,  $b(x)/a(x)$ , and  $b(\theta)/a(\theta)$  for  $\theta$  between  $H_{\perp c}$  and  $H_{\parallel c}$  and  $0 \leq x \leq 0.15$  for  $H_{\perp c}$ ; (b) for comparison, ESR spectra at 4.2 K and  $H_{\perp c}$  for single crystals grown in In and Zn fluxes and for the In-flux crystals of Ref. 6. Inset:  $\Delta H(T) = a + bT$  for Zn-flux crystals and  $H_{\perp c}$  at  $X$  and  $Q$  bands.

values increase with increasing  $x$  for  $T < 12\ K$ . At higher  $T$ , the effective  $g$  values are asymptotically  $T$  independent with a value of  $\approx 3.6$ . In the collective-mode description, the limiting value of  $g$  is identified with the  $Yb^{3+}$  Kramers doublet  $g$  factor, and the onset of the mode is associated with the decrease in the effective  $g$  value below 12 K to a value that lies between the  $ce$  and the  $Yb^{3+}$ . This decrease is less for  $x > 0$ , indicating that the disorder weakens the effect of the  $Yb$ - $ce$  correlations that give rise to the shift. General theoretical arguments<sup>25</sup> show that the effective  $g$  value of the collective mode is the weighted average of the  $g$  values of the  $ce$  and the  $Yb^{3+}$  ions. If one treats the interaction between the  $ce$  and the ions in a molecular-field approximation with interaction parameter  $\gamma$ , one obtains an expression for  $g_{\text{eff}} - g_{Yb}$  of the form  $g_{\text{eff}} - g_{Yb} = -(g_{Yb} - g_{ce})(g_{Yb}/g_{ce})\gamma\chi_{ce}$ , where  $\chi_{ce}$  denotes the susceptibility of the  $ce$ . Since it is unlikely that  $\chi_{ce}$  has the same temperature dependence as the measured susceptibility  $\chi_{\perp c}(T)$ , which is dominated by the contribution from the  $Yb^{3+}$  ions, we conclude that the interpretation of the observed  $g$  shift as arising from  $Yb^{3+}$ - $Yb^{3+}$  interactions is correct.

Figure 5(a) shows the  $a(x, \theta)$  data from Fig. 3. Usually, nonmagnetic impurities cause an inhomogeneous broadening of the ESR line ( $a \propto x$ ), which is in general attributed to a crystal-field distribution (CFD).<sup>17,26</sup> Natural impurities

and/or crystal defects also contribute to a CFD. As we mentioned, the broadening due to a  $c$ -axis misorientation of  $\leq 0.015^\circ$  cannot account for the observed increase in  $a$  near  $H_{\parallel c}$ . Therefore, we attribute this to an inhomogeneous broadening described by a distribution of  $g$  values  $\sigma_g(x) = \sigma_g(0) + \sigma'_g(x)x$  caused by a CFD. Then, we can write  $a(x) = a(0) + \sigma'_g(x)xH_{r\perp c}/g_{\perp c}$  and  $a(\theta) = a(0) + \sigma_g(0)H_r(\theta)/g(\theta)$ . From our data we obtain  $a(0) \approx 29\ Oe$ ,  $\sigma'_g(x) \approx 0.0114/\%Lu$ , and  $\sigma_g(0) \approx 0.0137$ . Notice that for  $\theta \approx 80^\circ$  and  $x \approx 0.15$  these broadenings are comparable. Figure 5(b) presents a comparison between the  $Yb^{3+}$  ESR X-band spectra at 4.2 K and  $H_{\perp c}$  for single crystals grown in In and Zn fluxes. The inset displays  $\Delta H(T) = a + bT$  and the fitting parameters for the crystal grown in Zn flux at  $X$  and  $Q$  bands and  $H_{\perp c}$ . The large  $a$  and  $b$  values are indications that Zn impurities were incorporated in the crystals. X-ray fluorescence measurements yield Zn concentrations of about 1%.

Our experiments confirm an ESR in  $YbRh_2Si_2$  below  $T_K \approx 25\ K$ .<sup>6</sup> 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 resemble the bottleneck and/or dynamic scenario for diluted magnetic moments exchange coupled to the  $ce$ , both with  $g \approx 2$ , where in a normal metal the SLR rate  $b$  and  $g$  value depend on the competition between the Korringa and Overhauser relaxation and the  $ce$  SLR.<sup>16,17,26,27</sup> Then, the increase in  $b$  by the addition of nonmagnetic impurities to  $YbRh_2Si_2$  (Lu and Zn in Figs. 3(b), 5(a), and 5(b) and La in Ref. 10) may be associated to “opening” the bottleneck regime due to the increase in the  $ce$  spin-flip scattering. Hence, we can write  $b(x) = b(0) + b'(x)x$ .<sup>17,26</sup> From our data, we estimate  $b(0) \approx 8\ Oe/K$  and a  $ce$  spin-flip scattering cross section  $b'(x) \approx 1.4(Oe/K)/\%Lu$ . Moreover, the  $\theta$  dependence of  $b$  [see Fig. 3(a)] may be another manifestation of the bottleneck effect. We argue that associated to the increase in  $a \propto \sigma_g(0)H_r(\theta)/g(\theta)$  there will be a significant number of *detuned*  $Yb^{3+}$  ions that may slow down the Overhauser relaxation and contribute to open the bottleneck regime. Assuming  $b(\theta) = b(0) + b'(\theta)\sigma_g(0)H_r(\theta)/g(\theta)$  we estimate  $b(0) \approx 7.5\ Oe$  and  $b'(\theta)\sigma_g(0) \approx 0.0032\ Oe^{-1}$ . We should mention that to *open* the bottleneck via *detuned*  $Yb^{3+}$  ions [ $\propto \sigma_g(0)H_r(\theta)/g(\theta)$ ] is only possible when the  $g$  value of the Kramers doublet is strongly anisotropic. Notice that in our case, this effect becomes important for  $\theta \geq 60^\circ$ . Figure 5(a) shows, as expected, that  $b(x)/a(x) \approx b(\theta)/a(\theta) \approx 0.26(4)$  for our data, confirming that a  $g$ -value distribution  $\sigma_g(x)$  due to a CFD is responsible for the increase in  $b$  and  $a$  in  $YbRh_2Si_2$ .

Another striking result reported in Fig. 2 is the nonmonotonic  $H$  dependence of  $b$ ,  $a$ , and effective  $g$  value of the  $Yb^{3+}$ - $ce$  resonant collective mode. The main difference with the bottleneck scenario given above is that the increase in  $b$  at  $S$  and  $Q$  bands is not followed by a systematic increase in  $a$ . Admixtures via Van Vleck terms<sup>28</sup> may be disregarded because this contribution should scale with  $H$ . Therefore, we believe that the low  $H$  tunability<sup>1,3,34</sup> 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 subtly tune and allows the formation of the resonant collective mode. We attribute the absence of low  $H$ -dependent ESR results in previous reports<sup>6</sup> to the presence

of “extrinsic” impurities and/or Rh/Si defects<sup>13,29</sup> that increase the SLR ( $b$ ) broadening the resonance and hiding the low  $H$  dependence of the ESR parameters in the NFL phase (such as for our Zn-flux crystals).

For this material, the  $\text{Yb}^{3+}$ -*ce resonant collective mode* presents the strongest bottleneck regime (smallest  $b$ ) at  $H \approx 1900$  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.<sup>11,30</sup> Recent calculations by Abrahams and Wölfle<sup>31</sup> suggested that the ESR linewidth may be strongly reduced by a factor involving the heavy-fermion mass and quasiparticle ferromagnetic (FM) exchange interactions ( $m/m^*$ ) [ $1 - \tilde{U}\chi_{ff,H}^{+-}(0)$ ]. These results indicate that the estimation of the linewidth from the Kondo temperature  $T_K$  is an overestimation. 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  $\text{YbRh}_2\text{Si}_2$  compound because (i) it presents an AF  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  exchange interaction [although other works in literature have claimed in favor of the existence of FM fluctuation in  $\text{YbRh}_2\text{Si}_2$  (Refs. 34 and 33)] and (ii) samples with the same thermodynamic properties present quite different linewidths [see Fig. 5(b)]. Furthermore, the anisotropy in the ESR in  $\text{YbRh}_2\text{Si}_2$  reflects both single-ion crystal-field effects and the  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  and  $\text{Yb}^{3+}$ -*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  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  and  $\text{Yb}^{3+}$ -*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  $\text{Yb}^{3+}$ -*ce* coupling is dominated by

a scalar interaction between the  $\text{Yb}^{3+}$  ground-state doublet pseudospins  $S_{\text{ps}}$  and the spins of the conduction electrons  $s$  with the consequence that the total spin  $s+S_{\text{ps}}$  is approximately constant of the motion.<sup>25,32</sup> 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.<sup>31</sup> The bottleneck scenario for the  $\text{Yb}^{3+}$ -*ce resonant collective mode* may also explain the absence of  $\text{Yb}^{3+}$  hyperfine ESR structure.<sup>35</sup> Finally, we hope that our results will motivate further theoretical approaches to understand the dynamics of strong exchange-coupled magnetic moments of unlike spins and  $g$  values, as  $\text{Yb}^{3+}$  and *ce*, and explore the general existence of a *resonant collective mode* with a bottleneck and/or dynamiclike behavior.

#### IV. SUMMARY

In summary, this work reports low  $H$ -dependent ESR, below  $T_K \approx 25$  K, in the NFL phase of  $\text{YbRh}_2\text{Si}_2$  ( $T \lesssim 10$  K). It is suggested that the observed ESR in  $\text{YbRh}_2\text{Si}_2$  corresponds to a  $\text{Yb}^{3+}$ -*ce resonant collective mode* in a strong bottlenecklike regime, which is highly affected by the presence of impurities, defects, and CFD. The analysis of our data allowed us to give estimations for the  $\text{Yb}^{3+}$ - $\text{Yb}^{3+}$  exchange parameter  $J_{ff}$  and a lower limit for the  $\text{Yb}^{3+}$ -*ce* exchange parameter  $|J_{fcl}|$ .

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