Ytterbium ESR in a lattice with weak coupling: the case of YbPt$_2$Sn

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Abstract. We observed a well-defined Yb$^{3+}$ electron spin resonance (ESR) line in YbPt$_2$Sn. The single crystal ESR data show strongly anisotropic $g$ factors, being largest in the basal plane, in agreement with magnetization data. This proves the intrinsic nature of the ESR signal. Analysis of these results indicates the crystal electric field ground state to be the $\Gamma_7$ doublet. Although the Kondo and intersite interactions are at least one order of magnitude weaker than in the Kondo lattices YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$, the temperature dependence of linewidth and $g$ factors show qualitative similarities.

1. Introduction

Electron spin resonance (ESR) in Yb- or Ce-based Kondo lattices has attracted considerable attention in the last years. The ESR line of 4$f$ Kondo ions was long thought to be undetectable because of a far too large linewidth of the size of the Kondo energy scale $T_K$. The observation of a very sharp ESR line in the Kondo lattice (KL) YbRh$_2$Si$_2$ with $T_K \approx 20$ K came as a big surprise [1], and resulted in a paradigm shift. Several theoretical studies have been devoted to the origin of this unexpected line narrowing [2, 3, 4]. ESR experiments on further Yb and Ce lattices have shown the relevance of strong ferromagnetic (FM) correlations, of the Kondo interaction, and of the anisotropy of the $g$ factor [5, 6, 7] for the ESR observability. However, the case is far from being clear because not all FM correlated KL show an ESR signal (e.g. YbNi$_4$P$_2$ [8] or CeFePO [9]) and also not all FM correlated non-Kondo systems are ESR silent (like YbRh or YbCo$_2$Si$_2$ [5, 6]). The strength of the exchange coupling between the 4$f$ ions and the conduction electrons (CE) crucially determines the ESR relaxation. For a strong coupling a coupled 4$f$-CE resonance mode is expected [2, 10] whereas a weak coupling favors a resonance mode arising from an isolated and local 4$f$ spin [11, 12]. Therefore, the observation and study of ESR in further Yb or Ce systems is of strong interest. In this respect it is worth to note the observability of a Ce-resonance in the Kondo lattice system CeB$_6$ [13]. However, the properties of this resonance seemingly do not match with expectations based on bulk measurements. The observability of this resonance and its very peculiar properties was proposed to originate from the rather unique antiferro-quadrupolar order observed in the cubic CeB$_6$[14].

The new compound YbPt$_2$Sn with a hexagonal ZrPt$_2$Al structure type has proven to be a metal with a very weak Yb-CE coupling and a paramagnet down to 0.25 K [15]. Susceptibility,
magnetization, resistivity, and specific heat measurements evidence a stable trivalent Yb state, without significant Kondo interaction. Furthermore, analysis of the magnetic entropy indicate intersite interactions to be remarkably weak, of the order of 0.8 K. Accordingly, specific heat data suggest short range ordering at about 250 mK. Regarding the above mentioned questions to the origin of ESR in Yb-lattice metals the extraordinary weak Yb-CE coupling (being much weaker than in YbRh or YbCo$_2$Si$_2$) qualifies YbPt$_2$Sn as an interesting compound for ESR investigations.

2. Experimental
In an electron spin resonance experiment an external, quasi static magnetic field $H$ tunes the Zeeman splitting of a spin state into resonance with a transverse microwave magnetic field. In case of resonance a net absorption of microwave power $P$ is observed which then contains the transverse dynamic spin susceptibility of the sample at microwave frequencies. 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 $dP/dH$. The ESR experiments were performed at X-band frequencies ($\nu \approx 9.4$ GHz) with a standard continuous wave spectrometer. The sample temperature was varied by using a He-flow cryostat.

We used a single crystalline platelet of YbPt$_2$Sn ($0.8 \times 0.8 \times 0.3$ mm$^3$) with a small residual electrical resistivity of about 200 $\mu$Ω cm. Sample preparation and characterization can be found in Ref. [15]. The crystal was mounted in the microwave cavity such that the microwave magnetic field was always within the hexagonal basal plane, probing the material within a penetration depth of $\approx \tau \mu$m.

The obtained absorption lineshape was described with a Lorentz form containing a considerable dispersion contribution as can be expected from the skin effect [11]. In addition, as the linewidth is comparable to the resonance field, it was necessary to take also into account the counter rotating component of the linearly polarized microwave field. Therefore a second Lorentzian at the negative resonance field contributes to the total lineshape [16].

3. Results and Discussion
For temperatures up to $\approx 16$ K we obtained absorption spectra containing well-defined ESR signals of YbPt$_2$Sn, see Fig. 1 for examples at 5 K. The resonance has a relatively large linewidth which hampered a proper extraction of the resonance parameters. For the field orientation $H \perp c$ the analysis could be considerably improved by taking advantage of the strong orientational dependence of the resonance field when rotating the crystalline $c$-axis in the external field. The ESR signals for $H || c$ were observed in an extended field range up to 1.8 T and could just be analyzed for temperatures below 8 K because of dominating noise and background.

As illustrated in Fig. 1 for $T = 5$ K the spectrum for $H || c$ is suitable to be used as a background spectrum: The ESR signal for $H || c$ of YbPt$_2$Sn is located at a resonance field of 0.95 T ($g_l = 0.7$), has a linewidth of $\approx 1$ T, and has about half of the amplitude of the $H \perp c$ signal. Thus, the background structures near zero and 0.3 T dominate the $H || c$ spectrum. As shown by the lower spectrum in Fig. 1, we therefore used the difference spectra $dP/dH(H \perp c) - dP/dH(H || c)$ for fitting the $H \perp c$ line. This was done by a Lorentzian shape with dispersion and absorption contributions in equal parts (which is reasonable because the penetration depth is much smaller than the sample thickness [11]).

The ESR $g$ factor, as determined from the resonance field $H_{res}$ of the Lorentzian line, $g = \hbar \nu / \mu_B H_{res}$, shows a remarkably large anisotropy, $g_\perp / g_\parallel \approx 7$ ($T = 5$ K), when aligning the crystalline $c$-axis to the field $H$ either perpendicular, $H \perp c$, or parallel, $H || c$. Interestingly, this anisotropy roughly agrees with values observed for Yb(Rh$_{1-x}$Co$_x$)$_2$Si$_2$ [6]. As shown in the right frame of figure 1, upon rotation between $H \perp c$ and $H || c$, we observed a clear uniaxial anisotropy of ESR $g$ factor. The particular values of $g_\perp$ and $g_\parallel$ can be understood by
considering the local magnetic properties of a crystalline electric field (CEF) split Yb$^{3+}$ $J = 7/2$ multiplet. The ground state turns out to be a $\Gamma_7$ Kramers doublet composed of the $| \pm \frac{1}{2} \rangle J_z$ states, while the first excited CEF level is about 1.7 meV above. The saturation magnetization expected for this ground state agrees well with the measured magnetization at $\mu_0 H = 7$ T and $T = 1.8$ K: $M_{H\parallel c} = 0.51 \mu_B$/Yb and $M_{H \perp c} - M_{\text{VV}} = 2.8 \mu_B$/Yb where $M_{\text{VV}}$ denotes Van Vleck paramagnetism contributing $0.5 \mu_B$/Yb to $M_{H \perp c}$. With $\mu = g \mu_B S_{\text{eff}}$ ($S_{\text{eff}} = \frac{1}{2}$) the $g$ values determined from these magnetization values are $g_\perp = 1.02$ and $g_\parallel = 5.6$ which correspond well with the values obtained by ESR at $T = 5$ K, $g_{\text{ESR}}^\perp = 0.7 \pm 0.6$ and $g_{\text{ESR}}^\parallel = 5.1 \pm 0.2$ (see Fig. 1).

The $g$ factor shows a strong temperature dependence below 5 K rising up to values of $g_\perp = 5.6$ at 3 K (see Fig. 3) and, hence, the difference between the experimental and CEF-expected $g$-values (a so-called $g$ factor shift) is within experimental error.

In metals with a Pauli-like susceptibility a $g$ factor shift contains the effective exchange coupling between the ESR-active local moment and the conduction electrons [17]. The apparently small $g$ factor shift in YbPt$_2$Sn agrees well with the exceptionally weak Yb-CE exchange coupling which is indicated in the low-temperature magnetic, and specific heat properties (Curie-Weiss term close to zero, saturation magnetization close to single-ion expectation, dominance of small intersite interactions instead of Kondo interactions) [15].

The intensity of the ESR signal $I_{\text{ESR}}$ provides further information on the local spin susceptibility. In our case of a broad linewidth compared to the resonance field a double integration of the ESR spectra $dP/dH$ yields approximate values of this susceptibility. $I_{\text{ESR}}$ was then determined from this doubly integrated value taking into account the temperature dependent skin depth $\delta \propto \sqrt{\rho(T)/\nu}$ given by the electrical resistivity $\rho(T)$ [15]. Figure 2 shows $I_{\text{ESR}}$ as functions of the temperature (left and middle frame) and of the bulk static magnetic susceptibility measured at $\mu_0 H = 1$ T (right frame). The temperature dependence of $I_{\text{ESR}}$ nicely follows a Curie-Weiss law $\propto (T - \Theta_{\text{ESR}})^{-1}$ with $\Theta_{\text{ESR}} = (2 \pm 0.3)$ K (dashed lines in Fig. 2).

**Figure 1.** Left frame: representative ESR spectra of YbPt$_2$Sn at 5 K for two orientations of external magnetic field, $H \perp c$ and $H \parallel c$. The lower spectrum results from subtracting the background from the $H \perp c$ spectrum (see main text). Solid line shows a Lorentzian lineshape [16]. Right frame: Dependence of the ESR $g$ factor on the angle $\varphi$ between the crystalline $c$-axis and magnetic field direction. Solid line denotes uniaxial behavior: $g(\varphi) = \sqrt{g_\perp^2 \sin^2 \varphi + g_\parallel^2 \cos^2 \varphi}$ with $g_\perp = 5.1 \pm 0.2$, $g_\parallel = 0.7 \pm 0.6$ (at $T = 5$ K).
Figure 2. Left frame and inset: Temperature dependence of integrated ESR intensity $I_{\text{ESR}}$ (arb. units) with a Curie-Weiss law [dashed line $I_{\text{ESR}} \propto 1/(T - \Theta_{\text{ESR}})$, $\Theta_{\text{ESR}} = (2 \pm 0.3) \text{ K}$]. Right frame: Dependence of $I_{\text{ESR}}$ on the bulk susceptibility $\chi$ measured at 1 T. Dotted line indicates linear extrapolation towards $\chi_0 = (1 \pm 0.5) \cdot 10^{-6} \text{ m}^3/\text{mol}$.

This positive value of $\Theta_{\text{ESR}}$ provides evidence for a dominantly FM Yb-Yb exchange coupling. Susceptibility data of a powder sample also indicate FM correlations, albeit with a smaller value $\Theta_{\chi} = 0.08 \text{ K}$ [15]. As shown in the right frame $I_{\text{ESR}}$ indeed follows the bulk susceptibility. In the temperature range $5 \text{ K} < T < 17 \text{ K}$ the plot evidences a linear relation (dotted line) which however crosses the abscissa at a finite bulk contribution $\chi_0 = (1 \pm 0.5) \cdot 10^{-6} \text{ m}^3/\text{mol}$. This value agrees well with the ESR silent Van Vleck contribution $\chi_{\text{VanVleck}} = 0.52 \cdot 10^{-6} \text{ m}^3/\text{mol}$ deduced from magnetization data. The departure from the linear behavior at low temperatures is partially due to saturation of the bulk susceptibility in the measuring field of 1 T (being larger than the resonance field of $\approx 0.2 \text{ T}$).

In the presence of Yb-Yb interactions the anisotropy of the resonant susceptibility crucially affects the temperature dependences of $g$ factor and linewidth [18]. This has been shown for the Yb$^{3+}$ resonance in the KL systems YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$ [19, 20, 21] as well as in YbCo$_2$Si$_2$ (with $T_K \ll 1 \text{ K}$) [6]. For the latter $g(T)$ could be successfully analyzed by the following molecular magnetic field description of the Yb-Yb interaction with exchange anisotropy $\theta \parallel - \theta \perp$: $g_{\perp}(T) = g_{\perp}^0 \cdot \left(1 - \frac{\theta \parallel - \theta \perp}{T - \Theta_{\perp}}\right)^\frac{1}{2}$ (1) $g_{\parallel}(T) = g_{\parallel}^0 \cdot \left(1 + \frac{\theta \parallel - \theta \perp}{T - \Theta_{\parallel}}\right)$ (2)

As shown in the left frame of Fig. 3 the $g(T)$ data nicely follow this description (dashed lines) with the parameters $\theta \perp = 2.3 \text{ K}$ and $\theta \parallel = 2.15 \text{ K}$ and $g_{\perp}^0 = 5$ and $g_{\parallel}^0 = 0.9$. Note the consistency of the Curie-Weiss temperatures $\theta \perp$ and $\Theta_{\text{ESR}}$ (see Fig. 2) stressing the validity of a molecular field description.

The weak exchange couplings among the Yb ions and between Yb and conduction electrons indicate that the Yb ions act as local ESR probes in a metallic environment. Then one expects the linewidth $\Delta B$ to be determined by the Korringa relaxation process, where the local moments
Figure 3. Temperature dependence of ESR $g$ factor (as determined from the resonance field) and linewidth $\Delta B$. The dashed lines fit $g(T)$ using a molecular magnetic field description of Yb-Yb interactions according eq.(1). The linear behavior of $\Delta B(T)$ (dashed lines) indicates a Korringa relaxation of local Yb$^{3+}$ spins by conduction electron spins.

are scattered by the conduction electrons, which should lead to a linear dependence of the linewidth on the temperature [17]. Indeed, as shown in the right frame of Fig. 3 $\Delta B(T)$ can be characterized by a linear behavior (dashed line) with a slope $\delta \Delta B/\delta T$ of 11 mT/K (180 mT/K) and a residual value $\Delta B_0$ of 130 mT (500 mT) for $H \perp c$ ($H \parallel c$, respectively). For $H \perp c$, below $T = 5$ K a saturation followed by a slight increase is visible which may indicate strong spin fluctuations from short range magnetic interactions. These are seen as a $1/T^2$ behavior of specific heat data above an anomaly due to short range magnetic order at 0.25 K [15]. The relatively large value of $\Delta B_0$ arises from disorder related relaxation channels in accordance with the observed low residual resistivity ratio [15]. In addition there is an “intrinsic” magnetic disorder from the non-negligible nuclear hyperfine coupling [15] which results in three magnetically different Yb$^{3+}$ spins on a single crystallographic site.

4. Conclusions
The very weak exchange coupling between intersite Yb ions as well as between Yb spins and conduction electron spins [15] leads to a well defined ESR of Yb$^{3+}$ spins with local single ion properties. This is similar to the situation of the Eu$^{2+}$ ESR in Eu-based pnictides [22, 23]. Hence, the Yb$^{3+}$ spins in YbPt$_2$Sn serve as intrinsic probes to characterize the local moment dynamics. A linear temperature dependence of the linewidth is due to Korringa scattering of conduction electrons whereas short range magnetic correlations lead to additional broadening towards low temperatures. The temperature dependence of ESR intensity and $g$ factor can consistently be described with positive, slightly anisotropic Weiss temperatures of about 2 K. $g_\perp$ and $g_\parallel$ well agree with the values expected from the saturation magnetization.

Comparing these results with the ESR in Yb-based Kondo lattices like YbRh$_2$Si$_2$ it is worth to note the similar temperature behavior as well as similar ESR parameters in some cases (the linewidth parameters of the weakly Co doped YbRh$_2$Si$_2$ [6], for instance). In Kondo lattice systems the presence of ESR could not be understood without the Yb-lattice Kondo interaction which crucially supports a collective spin mode of Yb and conduction electron spins [2] and which enables the observation of a heavy electron resonance [24]. It is important to note there that one does not observe a pure Yb$^{3+}$ resonance but a coupled Yb$^{3+}$-conduction electron resonant collective mode [20]. This in contrast to the situation in YbPt$_2$Sn where a pure Yb$^{3+}$ ESR can
be observed because of the very weak Kondo coupling. Also, as in Kondo lattice systems [5],
the predominant ferromagnetic Yb-Yb coupling may be relevant for narrow lines.

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