Electron Spin Resonance of Ferromagnetic Kondo Lattice Systems

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Introduction

One major focus in condensed matter physics is the investigation of compounds in which strong correlations among the charge carriers cause unusual, not understood properties. In this respect the spin (magnetic) degrees of freedom of the electrons are of particular interest. Electron Spin Resonance (ESR) spectroscopy allows to investigate elementary magnetic excitations in a standard setup at energies of about 0.1 meV. The observation of a well-defined ESR signal below the Kondo temperature T_K in the heavy-fermion compounds $YbRh_2Si_2$ and $YbIr_2Si_2$ refuted a common believe that concentrated rare earth ions in Kondo-lattice intermetallic compounds would be ESR-silent in the Kondo regime [1, 2]. The signal showed distinct properties of a local Yb^{3+} 4*f* spin [3, 4] and, hence, should contain valuable microscopic information on the dynamical Kondo coupling to the conduction electrons. A detailed analysis of the ESR spectra lineshape [5] as well as an investigation of the effect of replacing Yb with non-magnetic La on the ESR properties [6] provided some important information about the role of conduction electron spin relaxation in the ESR response. However, the consequences of this signal for understanding the Kondo ion physics in general as well as the prerequisites for its observation remained unclear. A plausible explanation would be the inherent difference between Yb- and the apparently ESR silent Ce- based Kondo lattices, the former ones presenting a much stronger local character which allows for a narrow, observable ESR line. Another mechanism could be based on the presence of ferromagnetic fluctuations, since $YbRh_2Si_2$ and $YbIr_2Si_2$ seem to be unique cases of Kondo lattice systems close to a critical point with strong ferromagnetic (FM) fluctuations and a concomitant strongly enhanced spin susceptibility. The latter leads to a reduced ESR line broadening as was recently shown to hold also for the ESR in Kondo systems [7].

In order to provide experimental results and arguments which allow to discriminate between the above scenarios we investigated the ESR of the Ce*T*PO (*T* = Ru, Os) compound series. These materials are Ce-based Kondo lattice systems presenting a strong tendency toward ferromagnetism [8]. For CeRuPO a pronounced decrease of the electrical resistivity at temperatures below 50 K indicates the onset of coherent Kondo scattering whereas the reference compound LaRuPO neither shows signatures of the Kondo effect nor of magnetism of the Ru atoms [8]. The temperature and magnetic field dependences of magnetic susceptibility and specific heat provide evidence for FM order at T_C = 15 K. Thus, CeRuPO (T_K~10 K) seems to be a rare example of an *f*-electron based FM Kondo lattice [8]. In contrast, CeOsPO shows antiferromagnetic (AFM) order at T_N = 4.4 K, despite only minor changes in lattice parameters and electronic configuration. Therefore, CeRuPO and CeOsPO are ideally suited to study the difference between FM and AFM Kondo lattices.

We present ESR results on these new Ce-based systems and discuss the ESR of further Ce- or Yb-based compounds with either ferro- or antiferromagnetic exchange and different strength of the Kondo interaction. The results demonstrate that the above mentioned unexpected ESR observation in heavy fermion metals is restricted neither to Yb-based compounds nor to compounds close to a quantum critical point, nor to compounds with a strong Kondo interaction. Instead, they indicate that the observability of the ESR signal in a dense Kondo lattice is connected to the presence of FM correlations between the Kondo ions [9].

Experimental

We used polycrystalline samples of Ce*T*PO (*T* = Ru, Os) as well as single crystals of CeRuPO whose preparation and characterization was described earlier [8, 10]. The tetragonal crystal structure (*P*4/*nmm*) contains alternating layers of OCe_4 and TP_4 tetrahedra. ESR probes the absorbed

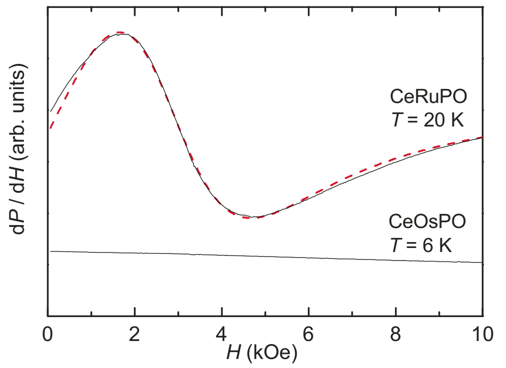

Fig. 1: X-band ESR spectra taken with same instrument settings for polycrystalline samples of CeRuPO (T_C = 15 K) and CeOsPO (T_N = 4.4 K). The red dashed line fits the data with a metallic Lorentzian line shape [9].

power P of a transversal magnetic microwave field as a function of an external, static magnetic field H. To improve the signal-to-noise ratio, we used a lock-in technique by modulating the static field. This yields the derivative of the resonance signal dP/dH. The ESR experiments were performed at X-band frequencies (ν ~ 9.4 GHz) with our Bruker ELEXSYS 500 spectrometer and at temperatures between 5 K and 125 K. The measured ESR spectra could be fitted with a Lorentzian lineshape. Here, we discuss the parameters which describe the lineshape: the linewidth (ΔH) and the resonance field (H_{res}). The latter determines an effective ESR g-factor ($g_{ESR}=h\nu/\mu_B H_{res}$). In case of a local ESR spin probe in a metallic environment, ΔH measures the spin-spin relaxation rate of the local spin, whereas g_{ESR} is determined both by the magnitude of the local moment and the static magnetic field at the spin site.

Results

Fig. 1 shows the main result of our ESR measurements on Ce*T*PO (*T* = Ru, Os) at temperatures just above the respective magnetic ordering temperature. The ESR investigations of both compounds were performed with the same instrument parameters and almost the same sample masses. For CeRuPO that shows FM correlations, a well-defined and intense ESR signal could easily be observed. On the other hand, the antiferromagnetically correlated CeOsPO is absolutely ESR silent, even when using the largest instrument amplifycation. The different nature of magnetic correlations in CeRuPO and CeOsPO [8] seems to be the only difference that is relevant for the ESR observation. In fact, many of their properties are similar: The preparation method is identical, the lattice parameters are very close, and the disorder effect on the electrical resistivity is comparable.

Besides the large signal-to-noise ratio and the large linewidth, the intrinsic origin of the ESR signal of CeRuPO is reflected by its anisotropy and temperature dependence being intimately related to the bulk static magnetic properties of the compound. Fig. 2 shows the variation of the ESR signal when the crystal is rotated in the magnetic field around an axis lying within the tetragonal basal plane. Both linewidth and resonance field of the ESR response exhibit uniaxial symmetry reflecting consistency with the crystal structure.

Fig. 3 displays the temperature dependences of the resonance field H_{res} and linewidth ΔH. The strongly anisotropic behavior of these quantities in CeRuPO reflects the collinear ferromagnetic correlations along the c-axis and an easy-plane, single-ion ground state as determined by the crystalline electric field (CEF) [10]. The temperature behavior of H_{res} is related to the effect of critical fluctuations on the internal magnetic field when an external field is applied. The critical fluctuations of the c-axis magnetization counteract the external field when it is applied in the basal plane ($H \perp$ c), i.e. the field required for resonance increases when T_C is approached from above. In contrast, if the external field H is applied along the c-axis ($H \parallel$ c) the growing internal field for $T \rightarrow T_C$ supports H, resulting in a suppressed H_{res}. The effect of critical

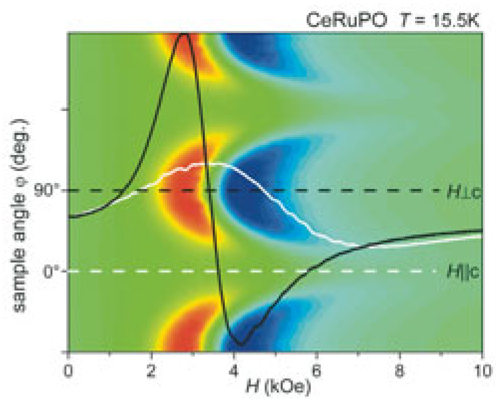

Fig. 2: Anisotropy of X-band ESR spectra of a single crystalline sample of CeRuPO (T_C = 14 K) illustrated by the color encoded spectra amplitudes at various angles φ between c-axis and external magnetic field. Two ESR spectra at φ = 0° (white) and φ = 90°(black) are shown to demonstrate the anisotropic linewidth and resonance field.

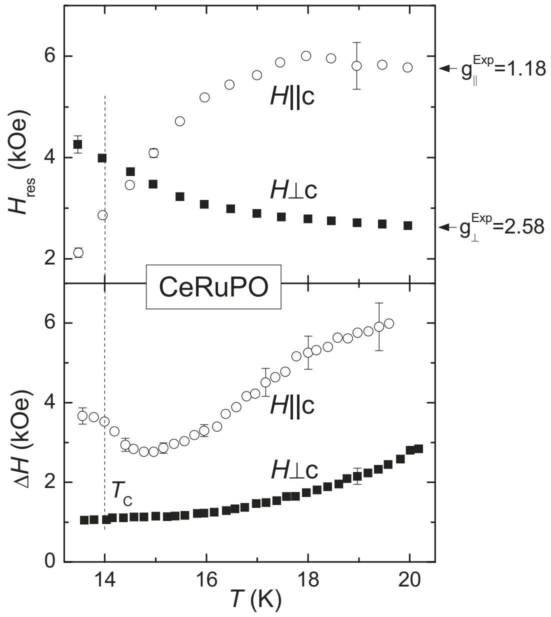

Fig. 3: Temperature dependence of the ESR resonance field H_{res} (top frame) and linewidth ΔH (bottom frame) for two c-axis orientations to the external magnetic field H. H_{res} (T > 20K) corresponds to the indicated g-factors, close to the values for a Ce^{3+} (Γ_6 doublet) state.

fluctuations is relevant for temperatures up to about 18K above which the observed resonance fields correspond to g factors ($g_{\perp,\parallel}^{Exp}$) which agree well with the Ce^{3+} CEF g-factors ($g_{\perp}^{CEF}$=2.57, $g_{\parallel}^{CEF}$ =0.86) (see Fig. 3). Both the temperature dependence and absolute values of the resonance field are highly consistent with the bulk magnetic properties of CeRuPO and, hence, can be taken as strong evidence for intrinsic Ce^{3+} spins being the source for the observed ESR signal.

The temperature dependence of the ESR linewidth ΔH (corresponding to the transverse spin relaxation) indicates a Korringa-type relaxation in a small T-region of the paramagnetic regime. The underlying mechanism of such a relaxation is a scattering of conduction electrons off the ESR-active spins leading to a linear-in-T behavior of ΔH. It is commonly observed for local spin probes that are doped into a metallic environment [11] and depends on the density of states at the Fermi level as well as the exchange coupling. In a metallic compound like CeRuPO (ρ_0 = 5 μΩcm) one expects the Korringa mechanism to provide the dominant relaxation. A different temperature dependence is observed for systems in which an isotropic spin exchange coupling effectively narrows the linewidth [12]. In case of CeRuPO, detailed investigations and analysis of the ESR anisotropy in the paramagnetic regime should provide relevant information about the contributions of Korringa and exchange-narrowing mechanisms. For temperatures around T_C the critical slowing-down of spin fluctuations reduces the effect of exchange-narrowing. Thus, the linewidth increases significantly for $H \parallel c$ because the external field stabilizes the FM, collinear order along the c-axis, see Fig. 3. For $H \perp c$ the applied resonance field of ~ 4 kOe is strong enough to stabilize the spin fluctuations and also the narrowing mechanism. Thus, no linewidth increase is observed. However, at L-band frequencies and $H \perp$ c, the resonance field (~ 0.2 kOe) is smaller than the internal field from the collinear order along the c-axis, i.e. in-plane spin fluctuations cannot be stabilized and the linewidth increases close to T_C (not shown).

Discussion

Our comparison of the ESR properties of Ce*T*PO (T = Ru, Os) provides a first evidence for a connection between the observability of the ESR line and the presence of FM correlations. We have obtained more experimental arguments for this conclusion by investigating other Yb or Ce based intermetallic compounds with either FM or AFM correlations and different strengths of the Kondo interaction (see Tab. 1). Besides the positive ESR observations in $YbRh_2Si_2$ and $YbIr_2Si_2$ which both exhibit strong FM correlations (as indicated, e.g., by an enhanced Sommerfeld-Wilson ratio) we found an ESR signal in another Yb-based compound with FM correlations, YbRh, which displays FM order of Yb^{3+} below T_C = 1.2 K. However, in contrast to the two above mentioned heavy-fermion compounds, Kondo-type features are found neither in the electrical resistivity nor in the specific heat of YbRh. We investigated further Ce- and Yb-based compounds with different strengths of the Kondo interaction, but all with dominant AFM correlations (see Tab. 1). Although the measurements were performed on high-quality single crystals in a wide temperature range around T_K and / or T_N, the magnetic ordering temperature, we did not observe an ESR signal in any of these systems. These observations provide overwhelming evidence for the importance of FM correlations for the narrowing of the ESR line – in remarkable analogy to the ESR in itinerant transition metal compounds. There, FM correlations may even enable an observable conduction electron spin resonance (CESR), as reported for $TiBe_2$ or $ZrZn_2$ (see Refs. in [9]). Recent theoretical considerations have proved the observability of an ESR signal in isotropic Kondo lattice systems with a Landau Fermi liquid ground state: A narrow linewidth results from 4f conduction electron hybridization and FM fluctuations [7]. For the FM Kondo lattice systems listed in Tab. 1 a putative heavy-CESR may be the origin of the observed ESR signal although its strongly anisotropic properties point to a local origin. Phenomenologically, the anisotropy may arise from the strong coupling between the local magnetic moments and the conduction electrons, and a "bottleneck" relaxation framework may apply as indicated from ESR experiments on $Yb_{1-x}La_xRh_2Si_2$ [6]. A corresponding formalism based on a local approach [4] and strong anisotropic exchange interactions is presently under preparation.

Compound	AFM	FM	KL	ESR signal
CeRuPO	-	√	√	Yes
CeOsPO	√	-	√	No
YbRh	-	√	-	Yes
$YbRh_2Si_2$	-	√	√	Yes
$YbIr_2Si_2$ (*I*-type)	-	√	√	Yes
$YbIr_2Si_2$ (*P*-type)	√	-	√	No
$Yb_4Rh_7Ge_6$	√	-	-	No
$YbNi_2B_2C$	√	-	√	No
$CeCu_2Si_2$ (S/A)	√	-	√	No
$CeNi_2Ge_2$	√	-	√	No
$CeCu_{6-x}Au_x$ ($x = 0,0.1$)	√	-	√	No

Table 1: Intermetallic compounds with predominant AFM or FM spin correlations investigated by X-band ESR [9]. "KL" denotes a Kondo lattice behavior in electrical resistivity or other properties.

Conclusion

A substantial amount of experimental data indicates the importance of strong FM correlations between the ESR-probed magnetic moments for the observability of an ESR signal in Kondo lattice systems. This conjecture is most strongly supported by the presence of an ESR signal in CeRuPO but its absence in CeOsPO. Here, the dominant magnetic interaction is switched from FM (CeRuPO) to AFM (CeOsPO) while keeping structural properties and disorder effects practically unchanged. These results provide a basic concept to understand the ESR in Kondo lattice systems and point out ESR as a prime method to investigate directly the spin dynamics of the Kondo ion. The standard ESR technique (X-band) is especially qualified for this purpose because the magnetic field (<10 kOe) is small enough to leave the Kondo state undisturbed in most compounds of interest. Detailed investigations on the field and temperature dependences of the ESR in $YbRh_2Si_2$ evidence a correlation between the evolution of the *g*-factor and the evolution of thermodynamic properties [13]. Such investigations should allow for a deeper insight into the formation of heavy quasiparticles in Kondo lattice systems.

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