Spin Resonance of Heavy Electrons: Investigation of Collective Spin Modes at High Pressures and Low Temperatures

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$YbRh_2Si_2$ and $YbIr_2Si_2$ belong to the few Kondo lattice compounds for which a well-defined Electron Spin Resonance (ESR) signal is observed and allows to directly characterize the spin dynamics of the Kondo ion. Even well below the (thermodynamically determined) single-ion Kondo temperature T_K this signal exhibits properties that are typical for localized 4*f* moments [1,2]. It has been shown that the presence of strong ferromagnetic correlations between the 4*f*-spins gives rise to a narrow, observable ESR signal [3,4], a situation which is very similar to the one for 3*d*-spins in ferromagnetic metals [5].

It is commonly believed that the occurrence of an ESR signal of paramagnetic ions as impurities in a metal is impeded by the Kondo effect. In this case, however, their magnetic moment should be screened by the conduction electrons at $T < T_K$ and their spin relaxation rate toward the conduction electrons (being in thermal equilibrium) should substantially exceed the ESR frequency. The fact that this is not always the case for Kondo lattice systems is a crucial issue of Kondo lattice physics, and was the main topic of our theoretical work [6,7] and extended supplementary ESR experiments [2-4, 8-10].

Theory development

The Kondo effect can help to observe an ESR signal in $YbRh_2Si_2$ – this was the central result of the developed theoretical understanding [7]. In the framework of this theory it also became clear that, in a simple approximation, the relaxation rate of Kondo ions diverges logarithmically for temperatures approaching T_K from above. However, it was found that this is correct only for a single-ion picture of the Kondo effect. In the case of a dense concentration of the paramagnetic impurities, and especially for the Kondo lattice, the back-influence of the Kondo ions on the spin dynamics of the conduction electrons becomes very important.

The equations of motion for both spin systems are coupled by additional kinetic coefficients, which diverge on the same energy scale: a *collective-spin-mode* is formed and is supported by the Kondo effect [7]. Mutual cancelation of all divergences results in a greatly reduced effective linewidth even in the case of a strongly anisotropic Kondo interaction. An additional reduction of the ESR linewidth results from the motional narrowing of the local magnetic field distribution caused by the quasi-localized *f*-electrons. This narrowing process is only possible if nearest-neighbor 4*f*-spins interact ferromagnetically. The importance of ferromagnetic fluctuations for narrow ESR lines is clearly supported by experimental results [3] and, noteworthy, was discussed also within a picture of a heavy-electron spin resonance [11].

As depicted in Figure 1 we were able to describe quantitatively the temperature and frequency dependence of the experimental data. It turns out that these data are consistent with a characteristic temperature of the Kramers doublet ground state that is much smaller than the usual Kondo temperature describing the 4*f*-thermodynamic properties.

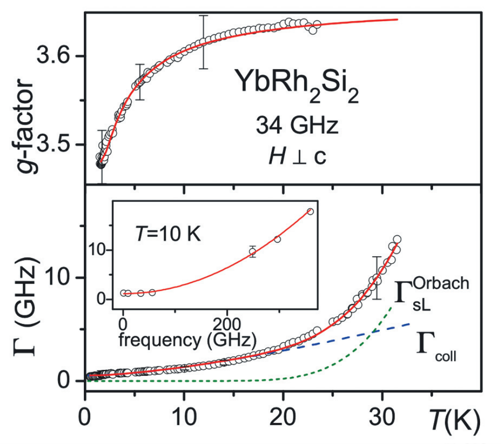

Fig. 1: g-factor and relaxation rate Γ of the ESR signal can be well described by a collective mode of Yb spins and conduction electron spins in the presence of the Kondo effect. Inset: frequency dependence of the collective mode rate. Solid and dashed lines are fits according to the “collective-spin-mode” scenario [7].

ESR of $YbRh_2Si_2$ under pressure and Co doping

The ESR investigations under external pressure in $YbRh_2Si_2$ were aimed towards tuning the 4*f*-electron hybridization. Consequently, these studies should provide additional information on the spin dynamics. The challenge of these experiments was the skin-depth limited, small ESR intensity of $YbRh_2Si_2$ and the need to measure at temperatures well below 20 K. Therefore, at the University of Augsburg, we extensively improved a previously home-built ESR pressure setup which allows for ESR measurements at the X-band (9.4 GHz) under a hydrostatic pressure of up to 3 GPa (30 kbar) and in a temperature range 2.5 K – 300 K [8].

Figure 2a shows examples of ESR spectra of $YbRh_2Si_2$ as a function of pressure. With increasing pressure the ESR line broadens (Fig. 2c) and shifts to higher fields, i.e. to correspondingly smaller *g*-values, (Fig. 2d). The present theoretical framework for the ESR in $YbRh_2Si_2$ [7, 11] provides a reasonable basis to understand the ESR under pressure in terms of the Kondo effect and the presence of ferromagnetic correlations. For instance, the concept of a narrowing of linewidth by the 4*f*-spin motion could provide, within the collective-spin-mode scenario [7], a preliminary understanding of the pressure-induced line broadening (Fig. 2c). An increase of pressure favors the magnetic Yb^{3+} configuration and, hence, seems to be equivalent to a slowing down of the 4*f*-spin motion resulting from a decrease in 4*f*-conduction spin hybridization.

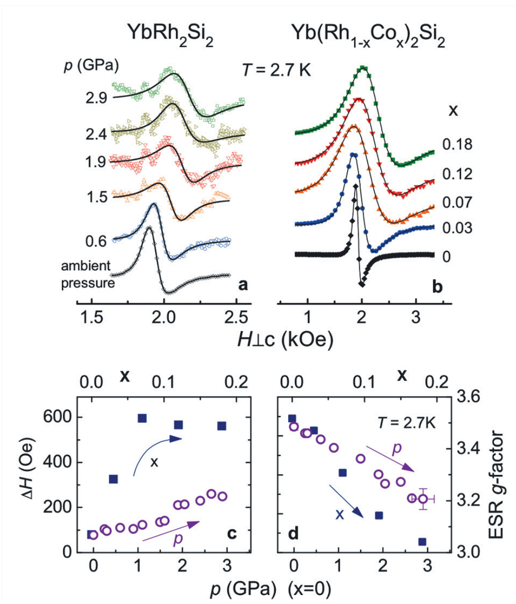

Fig. 2: ESR spectra dP/dH (P: absorbed microwave power) at 9.4 GHz with fitted metallic Lorentzian shapes (solid lines) (a) under different hydrostatic pressures *p* (amplitudes are scaled for best illustration), (b) at ambient pressure for different concentrations *x* of Co. Linewidth Δ*H*, (c), and effective g-factor, (d). Upper axis and lower axis refer to chemical and external pressure, respectively, and are related as described in the text.

We compared the pressure results with the X-band ESR of $Yb(Rh_{1-x}Co_x)_2Si_2$. Substituting the Rh ions with smaller Co ions causes a similar behavior of the ESR response (Fig. 2b). The volume reduction introduced by either external pressure *p* or internal "chemical pressure" *x* stabilizes antiferromagnetic ordering and gives rise to an equivalent evolution of the Néel temperature T_N. The *x* and *p* values are related according the measured lattice parameters and the bulk modulus of $YbRh_2Si_2$, and are shown at the ordinates of Figures 2c and 2d. There, for $p \geq 1$ GPa or $x \geq 0.07$, significantly different values of ESR linewidth and *g*-factor are revealed. Therefore, the static and dynamic magnetic properties of the Yb^{3+} spin seem to be influenced not only by the decrease of the unit cell volume but also by disorder induced by Co doping. The obtained ESR linewidths for all investigated pressures and Co contents could be characterized by a universal ratio between the respective residual linewidth and the slope of the linear temperature dependence of the linewidth. By relating both quantities to the scattering processes of charge transport the evolution of the ESR data with pressure allowed a further characterization of the influence of the Kondo interaction on the ESR of $YbRh_2Si_2$ [8].

The pressure dependence of the linear slope of the temperature evolution of the linewidth is equivalent to the linewidth behavior of local Gd^{3+} spins serving as diluted ESR probes in Ce-based heavy-fermion compounds [12]. There, the change in the low-temperature slope under pressure is explained by the change of the Kondo temperature. Therefore, the equivalency reveals two conclusions: (i) the ESR in $YbRh_2Si_2$ looks like a resonance of local Yb^{3+} spins in a metallic environment and (ii) the Kondo temperature is a relevant parameter to describe the linewidth.

ESR of $YbRh_2Si_2$ at low temperatures

Our ESR investigations of $YbRh_2Si_2$ at temperatures down to 0.5 K were aimed to investigate an eventual crossover behavior of the ESR parameters linewidth and *g*-factor once the electronic properties of $YbRh_2Si_2$ approach a heavy Landau-Fermi liquid behavior. This is seen, for instance, in the tempera-

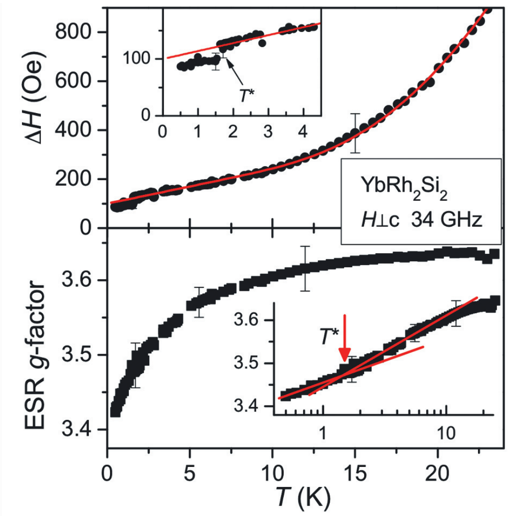

Fig. 3: Temperature dependence of the ESR linewidth ΔH and effective ESR g-factor. Temperature T^* indicates anomalies which occur at the boundary of heavy-electron formation. Red lines indicate linear and exponential behavior for ΔH and $\ln(T)$ for g [9].

ture dependence of the magnetic susceptibility which exhibits a maximum close to a temperature T^* specified by a crossover in the isothermal Hall resistivity.

The low-temperature ESR experiments were performed using home-built ^{3}He bath cryostats for X-band (University of Augsburg) and Q-band (Okayama University). For X-band (resonance field ≈ 2 kOe), the lowest accessible temperature of 0.69 K was still above T^*, and no anomalies were found in the ESR parameters [9]. The measurements at Q-band (resonance field ≈ 7 kOe) went down to 0.6 K and, thus, well below $T^* \approx 1.5$ K [9].

The data displayed in Figure 3 indeed establish a crossover behavior of the ESR linewidth and *g*-factor at $T \approx T^*$. At higher fields, although much weaker developed, such crossover behavior was also reported and interpreted as a typical signature of a heavy-electron spin resonance [10]. We suspect, however, that the anomaly of the ESR parameters at $T \approx T^*$ indicates a crossover for the resonance field defined by $k_B T^* = g\ \mu_B H_{res}$, i.e. for $T < T^*$ the resonance of the coupled 4*f*-conduction electron spins needs to be considered in a low-temperature limit (thermal energy falls below the Zeeman energy). We plan to develop a corresponding theoretical treatment in future.

Conclusion

We could show that the pressure effects on the spin resonance in $YbRh_2Si_2$ on the one hand manifest the local character of the Yb^{3+} spins, on the other hand indicate the properties of a collective 4*f*-conduction electron spin mode upon changing the 4*f*-conduction electron hybridization [8]. Furthermore, we found that both the ESR *g*-factor and the linewidth of $YbRh_2Si_2$ display a crossover behavior at around the same temperatures where thermodynamic and static magnetic properties indicate a crossover to Landau-Fermi liquid behavior [9].

A full treatment of these results within our recently developed theoretical approach of an anisotropic bottleneck scenario [7] will be the subject of future work.

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