Electronic hybridization effects in dense intermetallics measured by electron spin resonance

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Abstract. Recent theoretical studies associate the unexpected well resolved electron spin resonance (ESR) signals below Kondo temperature in several undoped heavy-fermion (HF) compounds with hybridization effects between $4f$ and conduction electrons (CE) in the presence of ferromagnetic (FM) fluctuations. We analyze ESR experiments in different concentrated Yb-, Ce-, and Eu-based intermetallic systems. We believe that the exotic ESR absorption here is caused by a novel type of ESR excitations – hybridized electronic states, which can be created in some strongly correlated electronic systems due to hybridization between the $4f$-orbitals and the wave functions of the CE of the outer electronic shells in conjunction with FM RKKY interaction.

1. Introduction
Recently, much attention has been focused on observation of well-defined ESR signals in several concentrated intermetallic systems [1-6] where the HF behaviour, evolution of the Kondo state, and hybridization mechanisms can be studied directly from the ESR measurements in contrast to earlier investigations in some Kondo lattices doped with different rare-earth ions as a spin probes [7]. According to several theories developed during two last years [8-12], an effective hybridization between localized $f$ electrons and conduction bands and strongly anisotropic Kondo-ion interactions in conjunction with FM correlations which reduce usually a very effective spin-lattice relaxation are discussed as a possible origins of the narrowed ESR lines below the Kondo temperature in these experiments. In the present contribution, we report the orientation-dependent ESR spectra in YbCo$_2$Zn$_{20}$ at room temperature and compare the results with those of all known ESR studies in dense intermetallic compounds.

2. Experimental results
Details of synthesis and characterization of the YbCo$_2$Zn$_{20}$ single crystals have been described previously [13]. The single crystal specimen used in our study has a size $5\times4\times4$ mm$^3$. ESR spectra of YbCo$_2$Zn$_{20}$ were recorded on an EMX/plus Bruker spectrometer operating at X-band. The strongly anisotropic ESR spectra with the resonance fields of the components up to 10 kOe have been detected at room temperature. Angular variations of the ESR lines are shown for two different orientations with the $dc$ external magnetic field $H$ rotated either in the $(10\bar{T})$ plane (Fig.1, top) or in the $(011)$ plane (Fig.1, bottom). For the latter orientation two resonances with different resonance field positions (at g

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~ 2.2 and g ~ 4.3) and line widths have been clearly resolved. Figure 2 illustrates the very different angular dependences of the effective ESR g-factor of both signals at this orientation.

Figure 1. Angular dependences of the ESR spectra at room temperature in the YbCo$_2$Zn$_{20}$ single crystal at 9.4 GHz for the constant magnetic field rotated in the (101) plane (top) and in the (011) plane (bottom).

3. Discussion

A coexistence of two ESR lines in YbCo$_2$Zn$_{20}$ reminds the results of recent ESR experiments in the first Yb-based HF superconductor $\beta$-YbAlB$_4$ where the single ESR response at g ~ 2.3 also at room temperature has been related to the CE spin resonance (CESR). At T = 4.2 K it reaches g ~ 3.0 and was attributed with the g-value of the Yb$^+$ Kramers doublet [14]. Although the signal at g ~ 2.2 in our studies can be ascribed to CESR too, the values of g ~ 4.3 for the low-field signal in YbCo$_2$Zn$_{20}$ and g ~ 7.33 in the iso-structural YbFe$_2$Zn$_{20}$ alloy [5] significantly differ from the corresponding quantities for the Yb$^{3+}$ ion. The average ESR g-factors and a possible hybridization types of all concentrated
intermetallic systems in which the ESR signals were observed are listed in the Table 1. For most compounds, these g-values are very close to those found for the corresponding 4f-ions Yb, Ce or Eu in different crystal symmetries.

![Graph showing the angular dependences for lines with g ~ 2.2 (full squares) and g ~ 4.3 (empty circles) of the ESR spectrum in the YbCo$_2$Zn$_{20}$ single crystal at room temperature. The external magnetic field has been rotated in the (011) plane.]

In contrast, in a case of the 4f-3d hybridization, i.e., for YbCuAl [15] and YbT$_2$Zn$_{20}$ (T = Co, Fe) the average ESR g-values can be attributed with those of the corresponding 3d-ions Cu, Co, and Fe. This means that the fraction of the 4f-electrons that hybridize with the CE and become itinerant can be much higher for the 4f-3d hybridization than for other types. At the same time, a part of 3d-electrons can be localized and the ESR g-tensor is given by the crystalline field scheme of the d states. Very recently, such coexistence of the 3d local moments and itinerant electrons in iron pnictides [17] and interplay of 3d and 4f magnetism in CeCoPO [18] have been discussed. In the latter compound the main contribution to magnetism results from the 3d-electrons of Co because the Ce 4f-electrons are

| Compound      | Average ESR g-factor | Hybridization type |
|---------------|----------------------|--------------------|
| YbCuAl        | 1.94$^{[15]}$        | 4f-3d3s3p          |
| YbCo$_2$Zn$_{20}$ | 2.2; 4.33$^{[5]}$   | 4f-3d4s           |
| YbFe$_2$Zn$_{20}$ | 7.33$^{[5]}$        | 4f-3d4s           |
| YbRh$_3$Si$_2$  | 3.55$^{[1,2]}$      | 4f-4d3p           |
| YbRh           | 2.55$^{[3]}$         | 4f-4d5s           |
| YbRh$_3$Pb     | 3.75$^{[4]}$         | 4f-4d6s6p         |
| CeRuPO         | 2.6$^{[5]}$          | 4f-4d2s2p         |
| YbIr$_3$Si$_2$  | 3.357$^{[3]}$       | 4f-5d3p           |
| YbBiPt         | 3.75$^{[5]}$         | 4f-5d6s6p         |
| $\beta$-YbAlB$_4$ | 2.3 (295 K); 3.0 (4.2 K)$^{[14]}$ | 4f-2s2p          |
| EuB$_6$        | 2.0$^{[6]}$          | 4f-2s2p           |

Table 1 Average ESR g-factors and possible hybridization types in some undoped intermetallic compounds investigated by ESR
screened due to pronounced Kondo-interactions. The exotic ESR response in dense intermetallics has been called as “HF ESR” [8, 19] or “Kondo coupled resonant modes” [9]. However, these ESR signals are not special to the HF systems or to the Kondo effect because they were also detected in the common alloys such as YbRh$_2$Pb [6] and EuB$_6$ [16] or in the non-Kondo lattice YbRh [4]. Up to now is not clear that either heavy mass only or/and the anisotropic part of the strong electron-electron interactions cause an observable ESR [11, 19]. Therefore, this novel type of ESR excitations can be treated as hybridized electronic states which arise in some strongly correlated electron systems in conjunction with FM correlations and lead to a narrow collective spin mode of coupled localized and itinerant electrons. Usually it is assumed that based solely on the ESR $g$ values and their anisotropy, we cannot distinguish between localized and conducting states resonating [9, 12]. Our orientation-dependent ESR measurements in YbCo$_2$Zn$_{20}$ provide probably the first experimental evidence of such separation.

In summary, we have discussed recent investigations on the ESR in dense intermetallic materials. A coexistence of CESR and local moment ESR signals has been observed in the strongly anisotropic ESR spectra of the YbCo$_2$Zn$_{20}$ crystals. The observed ESR behavior originates from the Co 3$d$-electrons rather than the Yb 4$f$-electrons due to very effective 4$f$-3$d$ hybridization.

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