

Electron Spin Resonance in the Heusler Alloy YbRh₂Pb

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An electron spin resonance (ESR) signal was observed in a concentrated Kondo lattice, Heusler alloy YbRh₂Pb. It is attributed to the combined effect of the 4*f* local magnetic moments of Yb³⁺ and conduction electrons. It is shown that the significant broadening and disappearance of the ESR line at temperatures above 20 K is caused by the processes of the spin-lattice relaxation of the Yb³⁺ ions through the first excited Stark doublet with an activation energy $\Delta \approx 73.5$ K. A comparison of the ESR data for YbRh₂Pb and some other undoped intermetallic compounds based on ytterbium, cerium, and europium indicates that *hybridized electronic states* occurring as the result of hybridization between the localized 4*f* electrons and the collectivized conduction electrons constitute a fundamentally new source of ESR.

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The interest in studies of heavy fermion (HF) compounds based on uranium, cerium, ytterbium, and other 4*f* and 5*f* elements has significantly increased recently. The magnetic properties, unique in many cases, of such concentrated Kondo systems are determined by the competition of several interactions of different natures (crystalline electric field (CEF), indirect interspin RKKY interaction, hybridization between *f* orbitals and the band states, screening of the localized magnetic *f* moments due to the Kondo effect, etc.) [1]. Using the electron spin resonance (ESR) technique to study these compounds is usually impossible without the introduction of additional paramagnetic probes (as a rule, rear-earth ions with a concentration of less than 2%), due to very intense spin-lattice relaxation of the *f* ions and the resulting large broadening of the ESR lines that cannot be resolved using standard ESR spectrometers. The observation of ESR signals below the Kondo temperature T_K in two undoped ytterbium-based HF metals, YbRh₂Si₂ and YbIr₂Si₂, pointed to a principally new possibility of using ESR for the direct study of HF compounds [2, 3]. The origins of these signals have not yet been understood and are intensely discussed [4–10]. Recent theoretical works [7, 8] attribute the ESR absorption in undiluted Kondo systems to the hybridization between the 4*f* electrons and the conduction electrons taking place in a system with the ferromag-

netic fluctuations that lead to the significant narrowing of the resonant line and thus make it possible to detect it. The mechanisms of such hybridization are as of yet poorly understood. In this paper, we report on the observation of ESR in another, recently synthesized, intermetallic compound YbRh₂Pb and analyze its spin dynamics as compared to two other similar HF compounds YbRh₂Si₂ and YbIr₂Si₂.

The compound YbRh₂Pb belongs to the group of triple Heusler alloys with a stoichiometric ratio of X₂YZ, where X and Y are transition metals and Z is an element of the III–V groups. The studied samples fabricated using the technique described in detail in [11] were small (2 × 1 × 0.5 mm) crystals. The X-ray structural analysis of the samples indicates the twinning of their crystal structure, the tetragonal symmetry of the unit cell, and inclusions of Pb (less than 1%) and smaller fractions of RhPb₂. In a temperature range of 100–300 K, the paramagnetic susceptibility χ followed the Curie–Weiss law with a Weiss temperature of $\Theta = -1.9 \pm 0.1$ K and the effective moment $M_{\text{eff}} = 3.3 \pm 1 \mu_B$ [11]. At $T < 20$ K, the temperature dependence of $1/\chi$ deviated from the linear law and antiferromagnetic ordering occurs below $T_N \approx 0.57$ K. The ESR measurements were made with a Bruker ESM/plus spectrometer (frequency ~ 9.45 GHz) in magnetic fields up to 1.4 T.

A single ESR signal with a distorted Dyson line-shape was observed in a temperature range from 4 to 25 K (Fig. 1). The g factor $g \sim 3.4$ obtained at $T = 5$ K turned out to be typical for Yb^{3+} ions in a tetragonal crystalline field [2], but the twinning of the sample prevented the detailed investigation of the anisotropy of the local paramagnetic center. The ESR line width ΔH_{pp} varied from 500 to more than 2000 Oe, i.e., in approximately the same range as for the YbRh_2Si_2 compound [2, 12]. However, its intensity was about 30 to 40 times smaller (for samples of approximately the same weight and size under identical signal detection conditions) and was comparable to the intensity of a background (parasitic) signal of the microwave cavity (the signal marked by the arrow in Fig. 1). The temperature dependence of ΔH_{pp} (Fig. 2) was described by the formula

$$\Delta H_{pp} = A + BT + C \exp(-\Delta/T), \quad (1)$$

where $A = 424 \pm 15$ Oe, $B = 27 \pm 2$ Oe/K, $C = 69.5 \pm 2$ kOe, and $\Delta = 73.5$ K.

The parameter A is determined by spin–spin interactions and inhomogeneous ESR line broadening. The term BT is the contribution to ΔH_{pp} from the Korringa relaxation caused by the exchange interaction of the spin of the ytterbium ion with the thermal fluctuations of the spin density of collectivized electrons. The third term in Eq. (1) is the result of thermal fluctuations of the electronic states of the Yb^{3+} ions, which lead to random transitions of these ions from the ground state to the first excited Stark sublevel with an activation energy $\Delta = 73.5$ K due to magnetic dipole–dipole and exchange interactions with the environment of these ions. This mechanism proposed in [2] to explain the temperature dependence of the parameters of the ESR spectrum of YbRh_2Si_2 is also responsible for the temperature dependence of the g factor (Fig. 3) at $T > 15$ K given by the expression

$$g(T) = g_0 + \Delta g_0 \exp(-\Delta/T), \quad (2)$$

$$\Delta g_0 = g_{\text{exc}} - g_0,$$

where g_0 and g_{exc} are the effective g factors of the ground and the first excited sublevels of the Yb^{3+} ion. Functional dependence (2) is shown in Fig. 3 as a solid line with $\Delta g_0 = -18.5 \pm 1.5$ and $g_{\text{exc}} = -15.1 \pm 1.5$. Thus, the temperature dependencies of the ESR parameters are typical for the properties of HF compounds: a transition from the mixed correlated state of localized $4f$ states of Yb and Fermi $3d$ electrons of Rh to a paramagnet with the magnetic moments of the $4f$ shells close to the magnetic moments of free Yb^{3+} ions [13]. The physical properties of the HF systems are determined by two different interactions: indirect exchange RKKY interaction and hybridization, i.e., the mixing of the wavefunctions of localized $4f$ electrons and collectivized electrons from the unfilled d , s , and p shells.

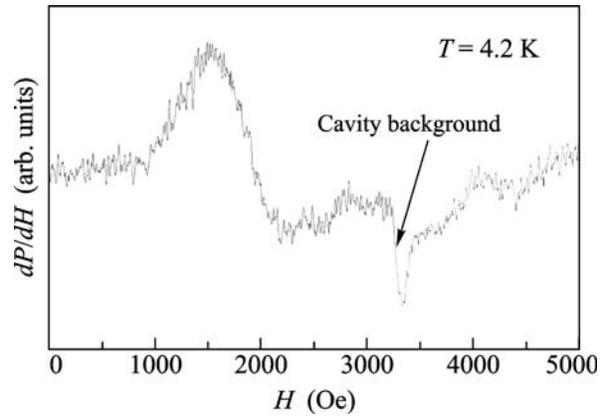


Fig. 1. Derivative of the absorption ESR signal in an YbRh_2Pb single crystal at 4.2 K. The arrow indicates the parasitic signal from the microwave cavity.

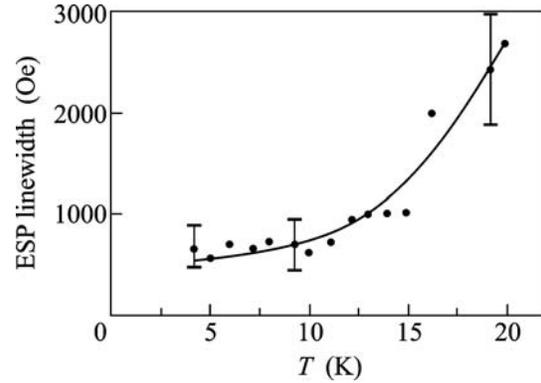


Fig. 2. Measured temperature dependence of the ESR linewidth in YbRh_2Pb at a frequency 9.45 GHz. The solid line presents formula (1) with the parameters $A = 424$ Oe, $B = 27$ Oe/K, $C = 69.5$ kOe, and $\Delta = 73.5$ K.

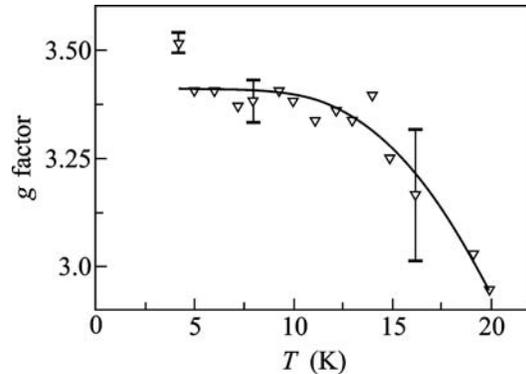


Fig. 3. Temperature dependence of the g factor obtained from the ESR experimental data at a frequency 9.45 GHz. The solid line is the theoretical calculation by Eq. (2).

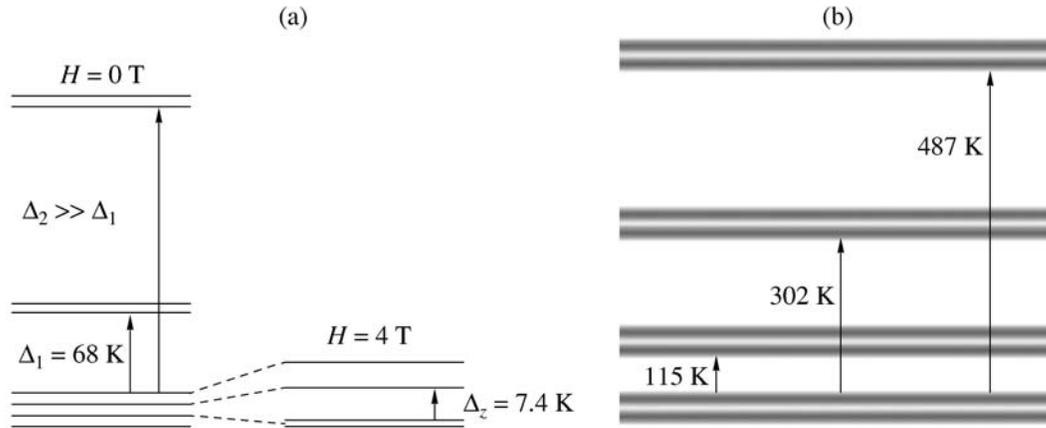


Fig. 4. Splitting of the main multiplet of the Stark structure of the Yb^{3+} ion in (a) YbRh_2Pb (according to data of [11]) and (b) YbRh_2Si_2 (calculated in [16]).

However, the electronic and magnetic properties of the Heusler alloys X_2YZ are primarily determined by both X – Y hybridization and the type of the sp element Z [14]. At the same time, the extent of such hybridization (the value of the overlapping of the wavefunctions) significantly depends on the chemical composition. This is why a comparison of the ESR experiments in YbRh_2Pb , YbRh_2Si_2 [2, 12], and YbIr_2Si_2 [3] makes it possible to estimate the probable f – d – p -hybridization effects. For example, the substitution of iridium Ir (with a configuration of the outer electron shells $[\text{Xe}]5d^76s^2$) for rhodium Rh ($[\text{Kr}]4d^85s^2$) (i.e., transition from YbRh_2Si_2 to YbIr_2Si_2) results in a significant decrease in the contribution of the collectivized electrons to the processes of the ESR relaxation, but does not significantly affect the ESR signal intensity. This is assumingly due to the weakening of the f – d hybridization owing to the decrease in the number of d electrons on the outer $5d$ shell of iridium. On the contrary, the replacement of silicon Si ($[\text{Ne}]3s^23p^2$) by lead Pb ($[\text{Hg}]6p^2$) does not result in a qualitative change of the spin dynamics in YbRh_2Pb as compared to YbRh_2Si_2 , since the f – d hybridization remains almost unchanged in this case (if one neglects the small change in the size of the unit cell and Yb – Rh interatomic distances [11, 15]). On the other hand, the extent of f – p hybridization in YbRh_2Pb can significantly decrease as compared to $4f$ – $3p$ hybridization in YbRh_2Si_2 due to the less efficient mixing of wavefunctions of the $4f$ and $6p$ shells, which is responsible for the strong suppression of the ESR signal intensity in the Pb-containing alloy.

The decrease in the extent of hybridization in YbRh_2Pb is also confirmed by the data on the determination of the location of the first excited Stark sublevel of the Yb^{3+} ion in YbRh_2Pb . The tetragonal symmetry of the crystal field for the Yb^{3+} ion ($4f^{13}$, $J = 7/2$) leads to the splitting of its multiplet into four Kramers dou-

plets [16]. Studies of the specific heat of YbRh_2Pb [11] showed that the two lowest doublets are located so close that they form a quadruplet in zero magnetic field, while the two other excited doublets are shifted from it by energies of $\Delta_1 = 68$ K and $\Delta_2 = 300$ K (see Fig. 4a). Thus, the Δ_1 value turned out to be very close to the energy interval Δ that was measured in our ESR experiments. In this respect, the situation with YbRh_2Si_2 was completely different: the Δ_1 value obtained from the neutron scattering spectra (Fig. 4b) was almost 1.5 times larger than the corresponding value, 115 K, obtained from the ESR measurements [2]. At the same time, it is well known that effective hybridization between the wavefunctions of localized $4f$ electrons and collectivized conduction electrons in the HF compounds leads to very wide (“smeared”) levels of the crystal field [17]. This smearing results in a large error in the determination of the location of the Stark sublevels of the RE ions from the ESR data for YbRh_2Si_2 and YbIr_2Si_2 as compared to neutron spectroscopy data [16]. In the usual compounds such as the YbRh_2Pb alloy, this error is relatively small and the measured Δ_1 value is smaller than the real one by no more than 15–20% [18].

Thus, the relatively small ESR absorption intensity in YbRh_2Pb can be explained by the very small extent of the f – p hybridization between the electronic states of the $4f$ -ion Yb^{3+} and the corresponding band electrons. Such a weak f – p hybridization most probably leads to a very small value of the RKKY interaction in YbRh_2Pb [11], which is responsible for the appearance of the ferromagnetic fluctuations primarily responsible for the observation of the ESR in concentrated HF systems [6, 7].

During the last year, several more observations of the ESR in different concentrated intermetallides were reported. The physical properties of these com-

pounds can be understood only with the inclusion of $f-d-s-p$ hybridization. They include not only HF systems (YbBiPt and YbCo₂Zn₂₀ [19]) or Kondo lattices (CeRuPO [6] and EuB₆ [20]), but also intermediate valence compounds YbFe₂Zn₂₀ [19] and the YbRh alloy [6] that does not exhibit the Kondo effect. In this case, the ESR signal is determined by the contributions of both the localized $4f$ moments and the magnetic moments of the collectivized electrons; these contributions cannot be separated by the ESR technique [8]. These facts indicate that hybridized electronic states occurring in undoped strongly correlated electron systems as a result of the competition of the ferromagnetic RKKY interactions and hybridization between the $4f$ -orbitals and the wavefunctions of the conduction electrons of the outer d , s , and p shells constitute a fundamentally new source of ESR. For this reason, the ESR technique can be used to study the hybridization effects in strongly correlated electron systems along with X-ray spectroscopy, neutron scattering, and angular resolved photoemission spectroscopy.

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