Quadrupolar interactions in heavy fermion metal YbRh$_2$Si$_2$

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Abstract

We describe the experimentally revealed by Sichelschmidt et al., Phys. Rev. Lett. 91 (2003) 156401, g tensor, $g_{||}=3.561$ and $g_{\perp}=0.17$, at 5 K by means of crystal field interactions of the $4f^{13}$ configuration of the Yb$^{3+}$ ion of YbRh$_2$Si$_2$ in a slightly orthorhombically distorted tetragonal crystal field. We have shown that the temperature dependence of the quadrupolar interactions $Q(T)$ of the Yb nucleus will help to distinguish between $\Gamma_7$ and $\Gamma_6$ ground state. For the $\Gamma_7$ ground state $Q(T)$ is expected to exhibit an anomalous dependence.

Key words: heavy fermion, crystal field, quadrupolar moment, YbRh$_2$Si$_2$

Recently Sichelschmidt et al. [1] reported the first successful Electron Spin Resonance (ESR) studies on single crystalline heavy fermion metallic compound YbRh$_2$Si$_2$ that allowed for the unambiguous observation of the localized $f$ states in a Kondo compound. The importance of this experiment relies in the fact, that practically all theories devoted to heavy-fermion phenomena were taking the itinerant or band behavior of $f$ electron as the starting point. Theoretical approaches to heavy-fermion phenomena with localized $f$ states were simply rejected both by referees of prestigious physical journals and by their editors that simply prohibited any discussion of the localized magnetism and crystal-field (CEF) interactions. Thus, the discovery by the Prof. F. Steglich’s group is really of great importance for theoretical understanding of heavy-fermion compounds. The authors of Ref. [1] are fully aware of the theoretical importance of their observation putting a lot of attention to evidence that this ESR signal comes out from the bulk unscreened Yb$^{3+}$ moments below Kondo temperature $T_K$. By means of the ESR experiment, at the external field $B=0.188$ T with the X-band frequency of 9.4 GHz, Sichelschmidt et al. [1] revealed the existence of a localized doublet characterized by very anisotropic $g$ tensor, with $g_{||} = 3.561$ and $g_{\perp} = 0.17$ at 5 K.

The aim of this paper is to report our search for the atomic-scale CEF parameters describing the observed $g$ tensor in order to find the microscopic origin of this localized state and its experimental verification.

We have attributed the observed state to the Yb$^{3+}$ configuration, more exactly to the strongly-correlated $4f^{13}$ configuration that, being in the Hund’s rule ground multiplet $^2F_{7/2}$, is described by $J = 7/2$. Our studies show that the higher multiplet $^2F_{9/2}$ does not affect the ground-multiplet properties. The crystal field of the tetragonal symmetry:

$$H_{CEF} = B^0_{2}O^2_2 + B^0_{4}O^4_4 + B^0_{6}O^6_6 + B^0_{4}O^4_0 + B^0_{6}O^6_0$$

splits the 8-fold degenerated multiplet $^2F_{7/2}$ into 4 Kramers doublets, $2\Gamma_6$ and $2\Gamma_7$. The lower in energy states take the form (notation by $\sin \alpha$ and $\cos \alpha$ assures the automatic normalization and the sign corresponds to 2 Kramers conjugate states):

$$\Gamma_6^1 = \sin \alpha |\pm 1/2 > + \cos \alpha |\mp 7/2 >$$
$$\Gamma_7^1 = \sin \beta |\pm 3/2 > + \cos \beta |\mp 5/2 >$$

We intend to reproduce the $g$ tensor keeping the overall CEF splitting $\Delta_{CEF}$ of size of 600-750 K (55-70 meV) and the first excited level at $D$ of 70-100 K (6-9 meV) in order to assure the proper thermodynamics and a reasonable magnitude of CEF interactions.

We have found that for the $\Gamma_7^1$ ground state the perfect reproduction of the ESR results, $g_{||} = 3.561$ ($J_{\perp} = \pm 1.56$) and $g_{\perp} = 0.17$ ($J_{||} = \pm 0.08$), is obtained for parameters: $B^0_{2} = +14$ K, $B^0_{4} = -60$ mK, $B^0_{6} = -0.5$ mK, $B^0_{4} = -2.30$ K and $B^0_{6} = -10$ mK with a small local orthorhombic distortion $B^0_{2} = +0.22$ K [2]. These
parameters yield the ground Kramers doublet:

\[ \Gamma_6 \cong \begin{bmatrix} 0.803 & 0.595 \\ 3/2 & 5/2 \end{bmatrix} \sim \begin{bmatrix} 1/2 & 7/2 \end{bmatrix} \]

that is characterized by \( J_{\perp} = \pm 1.560, J_{\parallel} = \pm 0.081 \) and \( Q_f = -4.7 \). The excited states are at 85 K (\( \Gamma_6^2 \) with \( Q_f = -13.8 \)), 485 K (\( \Gamma_7^2 \) and 688 K (\( \Gamma_7^2 \)). This anomalous dependence was discussed in Ref. [4].

For the \( \Gamma_6^2 \) 

\textbf{ground state}, attained for instance by tetragonal CEF interactions \( B_{1}^2 = +10 \text{ K}, B_{2}^0 = -75 \text{ mK}, B_{4}^0 = +7.5 \text{ mK}, B_{4}^2 = -2.22 \text{ K}, B_{6}^2 = -6.2 \text{ mK} \) with \( B_{2}^2 = +0.60 \text{ K}, the eigenfunction:

\[ \begin{align*}
\Gamma_6^2 &= 0.944 |\pm 1/2 > + 0.322 |\mp 7/2 > - 0.052 |\mp 3/2 > - 0.047 |\pm 5/2 > \\
\text{yields } J_{\perp} &= \pm 1.561, J_{\parallel} = \pm 0.084 \text{ and } Q_f \approx -11.2 \\
\text{for these parameters the excited states are at } 92 \text{ K (} \Gamma_7^1 \text{ with } Q_f = -3.4 \text{), 538 K (} \Gamma_7^2 \text{) and 546 K (} \Gamma_7^2 \text{).}
\end{align*} \]

Both sets equally well reproduce the 

g tensor and quite well the overall temperature dependence of the paramagnetic susceptibility \( \chi(T) \) and its huge anisotropy, presented in Fig. 1a of Ref. [3], the preference for the magnetic ordering with moments perpendicular to the \( c \) axis, the magnetization curve for external magnetic fields up to 60 T applied along the tetragonal \( c \) axis (Fig. 1b of Ref. [3] - the magnetization at 2 K and at 60 T amounts to 0.85 \( \mu_B \)). Inelastic-neutron-scattering results are not known yet so it is hardly possible to distinguish these states. We propose that they can be distinguished by the Mossbauer experiments, in particular by measurement of the quadrupolar moment \( Q_f \) of the 4f shell. As is shown in Fig. 1 the temperature dependence of \( Q(T) \) is completely different for both states. In case of the \( \Gamma_7^2 \) ground state \( Q_f \) shows a non-monotonic dependence. This anomalous dependence was discussed in Ref. [4] within the CEF-based model.

\[ Q(T) = Q_0 - \frac{T}{T_0} \]

\[ \text{Fig. 1. Temperature dependence of the quadrupolar moment of the } 4f \text{ shell in YbRh}_2\text{Si}_2 \text{ calculated for two different ground states } \Gamma_6 \text{ and } \Gamma_7. \]

We do not think that the present sets of CEF parameters are the final ones. There is a great number of sets that produce the shown ground-state eigenfunction (the simplest can be obtained by multiplication of all parameters by a constant positive value which causes equal spreading of the electronic structure) but surely the obtained sets substantially confine the searching area for CEF parameters. Though we worked hard by last 20 years in the evaluation of CEF and exchange interactions in different rare-earth compounds, both ionic and intermetallics [5,6] but the existence of so well-defined (so extremely thin) CEF states in a metallic compound YbRh_2Si_2 is a big surprise indeed. This compound was regarded as one of the prominent heavy-fermion compound with itinerant \( f \) electrons and with a substantial Kondo temperature (at least 25 K [1]). In the developed by us Quantum Atomic Solid State (QUASST) Theory [7,8], we recognize that the standard CEF approach is a gigantic correler electron approach to compounds containing open-shell transition-metal atoms. In our understanding the Kondo temperature is related to the energy of the first excited CEF doublet and the Kondo resonance is related to the removal of the Kramers degeneracy that is a source of low-energy, below 0.2 meV, excitations. These excitations are neutral, spin-like excitations and they are responsible for large low-temperature specific heat, a hallmark of heavy-fermion physics. Recently CEF states have been also revealed in another heavy-fermion metal UPd_2Al_3, that exhibits antiferromagnetism below 14 K and superconductivity below 2 K. These states are related to the \( 5f^2 \) configuration [6].

\textbf{In conclusion}, we have derived CEF parameters of the tetragonal symmetry with a small orthorhombic distortion that perfectly reproduce the ESR values (\( g_{\perp} = 3.561 \) and \( g_{\parallel} = 0.17 \)) as well as provide good reproduction of thermodynamical properties both for the \( \Gamma_6^2 \) and \( \Gamma_7^2 \) ground state. The proposed Mossbauer experiment for the evaluation of temperature dependence of the quadrupolar moment of the 4f shell can distinguish between these two ground states.

\textbf{References}

[1] J. Sichelschmidt et al, Phys. Rev. Lett. 91 (2003) 156401.
[2] R. J. Radwanski and Z. Ropka, Crystal field ground state in heavy fermion metal YbRh_2Si_2. cond-mat/0312725
[3] J. Custers et al, Acta Phys. Pol. B 32 (2001) 3211.
[4] R. J. Radwanski, J. Alloys-Compds 232 (1996) L5.
[5] R. J. Radwanski et al, J.Phys.: Condens. Matter 4 (1992) 8853.
[6] R. J. Radwanski, R. Michalski and Z. Ropka, Physica B 276-278 (2000) 803.
[7] R. J. Radwanski, R. Michalski, and Z. Ropka, Acta Phys. Pol. B 31 (2000) 3079.
[8] R. J. Radwanski and Z. Ropka, Quantum Atomic Solid State Theory, cond-mat/0010081.