Spin fluctuations in heavy-Fermion compounds YbZnCu$_4$ and YbAuCu$_4$, investigated by $^{63}$Cu NMR/NQR

Takehide Koyama$^a$, Takeshi Mito$^a$, Shinji Wada$^a$, John L. Sarrao$^b$,

$^a$ Department of Material Science, Graduate School of Science & Technology, and Department of Physics, Faculty of Science, Kobe University, Nada, Kobe 657-8501, Japan

$^b$ Los Alamos National Laboratory, Mail stop K 762, Los Alamos, NM 87545, USA

Abstract

We have investigated microscopically the heavy-Fermion properties of YbZnCu$_4$ and YbAuCu$_4$ with $^{63}$Cu NMR and PQR measurements. Both the isotropic and axial Knight shifts for each of the compounds showed a Curie-Weiss-type behavior, which is indicative of the localized Yb$^{3+}$ moments. The nuclear spin-lattice relaxation rate ($T_1T$)$^{-1}$ for YbZnCu$_4$ above 1.4K and for YbAuCu$_4$ above $\sim$ 50 K was proportional to the uniform susceptibility $\chi$, indicating that the correlation time $\tau_c^{-1}$ of Yb-spins is nearly independent of temperature. ($T_1T$)$^{-1}$ for YbAuCu$_4$ below $\sim$ 50 K exhibited a prominent increase associated probably with the decrease in $\tau_c^{-1}$ to the Kondo fluctuation rate $\tau_K^{-1} = k_B T_K/\hbar$.

Key words: heavy Fermion; spin fluctuations; Ytterbium cuprate;

The Yb$X$Cu$_4$ series ($X$=rate transition-metal elements) with cubic C15b (AuBe$_5$)-type crystal structure shows a wide variety of physical properties with the species of $X$ atoms. Among the compounds, YbAuCu$_4$ is a prototypical heavy-Fermion compound (electronic specific heat coefficient $\gamma$ $\sim$150 mJ/mol, Kondo temperature $T_K$ $\sim$ 2 K) and exhibits antiferromagnetic ordering below 1K [1]. While YbZnCu$_4$ ($\gamma$ $\sim$150 mJ/mol, $T_K$ $\sim$30 K) has been relatively less studied. The magnetic susceptibility $\chi$ of YbZnCu$_4$ showed a Curie-Weiss-type behavior at high temperatures with a ferromagnetic increase below $\sim$30 K. From the large upturn in the electronic resistivity at low temperatures, YbZnCu$_4$ has been suggested to be a failed Kondo semimetal [1].

In this paper, we report the results of the nuclear magnetic resonance (NMR) and pure quadrupole resonance (PQR) of $^{63}$Cu in YbZnCu$_4$ and YbAuCu$_4$, which can provide microscopic information on the static and dynamical properties of Yb spins.

1 E-mail:koyama@cphys.cla.kobe-u.ac.jp

Fig. 1. Temperature dependence of the isotropic and axial knight shift for YbZnCu$_4$ and YbZnCu$_4$.

The $^{63}$Cu NMR measurement was carried out under magnetic fields of $\sim$7 T with a phase-coherent pulsed
spectrometer operating at a constant frequency of 75 MHz. The NMR spectra exhibit the general electric-quadrupole split powder-pattern, and the values of the isotropic Knight shift \( K_{\text{iso}} \) and axial Knight shift \( K_{\text{ax}} \) are deduced from the spectrum analysis described in ref. [2]. For the temperatures below \( \sim 40 \) K, we could not obtain any reliable values of the Knight shift, because of the severe broadening of the line width. Fig. 1 shows the temperature dependence of \( K_{\text{iso}} \) and \( K_{\text{ax}} \) for YbZnCu\(_4\). The data for YbAuCu\(_4\) in the figure were cited from our previous report [2]. Both \( K_{\text{iso}} \) and \( K_{\text{ax}} \) for each of the compounds exhibit the Curie-Weiss type behavior, that can be ascribed to the localized Yb\(^{3+}\) moment. The Knight shift versus susceptibility plots are on a straight line, and the slope gives the value of anisotropic and isotropic terms of the transferred hyperfine coupling constants \( H_{\text{hf}}^{\text{iso}}(\text{tr}) \) and \( H_{\text{hf}}^{\text{ax}}(\text{tr}) \) as follows: -1.3 and -1.4 kOe/\( \mu_B \) for YbZnCu\(_4\); -0.97 and -1.2 kOe/\( \mu_B \) for YbAuCu\(_4\).

The spin-lattice relaxation rate \( T_1^{-1} \) of \(^{63}\)Cu was measured at peak intensity of the PQR line: 8.9 MHz for YbAuCu\(_4\); 12.6 MHz for YbZnCu\(_4\). Fig. 2 shows the temperature dependence of \( T_1^{-1} \) for each of the compounds. The data for YbAuCu\(_4\) are in good agreement with that reported by Nakamura et al. [3]. The \( T_1^{-1} \) data for each of the compounds do not obey the Korringa-like relation \( (T_1T)^{-1} = \text{const.} \), within the present experimental temperature range. Then we replotted in Fig. 3 the \( (T_1T)_{\text{tr}}^{-1} \) data against the corresponding \( \chi \) data. For the relaxation process to the fluctuating local moments with the correlation time \( \tau_f \), the relaxation rate is given by [4]

\[
(T_1T)_{\text{tr}}^{-1} = 2\gamma_n^2 k_B H_{\text{hf}}(\text{tr})^2 \frac{\chi}{\mu_B N} \tau_f, \tag{1}
\]

assuming a Lorentzian shape of fluctuation spectrum and \( \chi(q) \sim \chi(0) \). Here, \( \gamma_n \) is the nuclear gyromagnetic ratio, \( z \) the number of neighboring spins, and \( N \) the Avogadro’s number. The linear dependence of \( (T_1T)^{-1} \) on \( \chi \) for YbZnCu\(_4\) above 1.4 K and for YbAuCu\(_4\) above \( \sim 50 \) K indicates that \( \tau_f \) is nearly independent of the temperature. This is consistent with the strongly localized scheme for the 4f moments in these compounds. Taking the experimental values of \( (T_1T)^{-1} \) and \( H_{\text{hf}}^{\text{iso}}(\text{tr}) \) at high temperatures, we can estimate an order of \( f \) spin fluctuation energy \( T_f = h/k_B \tau_f \) as \( \sim 100 \) K for YbZnCu\(_4\) and \( \sim 20 \) K for YbAuCu\(_4\), respectively.

For YbAuCu\(_4\), \( (T_1T)^{-1} \) below \( \sim 50 \) K exhibited a prominent increase, and deviates from the linear dependence on \( \chi \). If we bravely use eq. (1) for the \( (T_1T)^{-1} \) data below \( \sim 50 \) K, though it is not really very satisfactory for the correlated spin system, \( \tau_f^{-1} \) decreases monotonously and approaches the order of 1 K. It is worth noting that \( \tau_f \sim 1 \) K can reasonably be compared with \( T_K \). The temperature independent \( \tau_f \sim 1 \) for YbZnCu\(_4\) down to 1.4 K suggests that the Kondo temperature for YbZnCu\(_4\) is much lower than \( T_K \sim 30 \) K estimated from the \( \chi \) data. The lack of a predominant Kondo compensation of 4f moments in YbZnCu\(_4\) down to \( \sim 1 \) K is considered to be consistent with the Kondo semimetal behavior observed in the resistivity.

References