Spin fluctuations in heavy-Fermion compounds $YbZnCu_4$ and $YbAuCu_4$, investigated by ^{63}Cu NMR/NQR

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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 ~ 50 K was proportional to the uniform suscsptibility χ, indicating that the correlation time τ_f^{-1} of Yb-spins is nearly independent of temperature. $(T_1T)^{-1}$ for $YbAuCu_4$ below ~ 50 K exhibited a prominent increase assocated probably with the decrease in τ_f^{-1} to the Kondo fluctuation rate $\tau_K^{-1} = k_B T_K/\hbar$.

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

The YbXCu$_4$ 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 γ ~150 mJ/mol, Kondo temperature T_K ~2 K) and exhibits antiferromagnetic ordering below 1K [1]. While $YbZnCu_4$ (γ ~150 mJ/mol, T_K ~30 K) has been relatively less studied. The magnetic susceptibility χ of $YbZnCu_4$ showed a Curie-Weiss-type behavior at high temperatures with a ferromagnetic increase below ~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 dymamical properties of Yb spins.

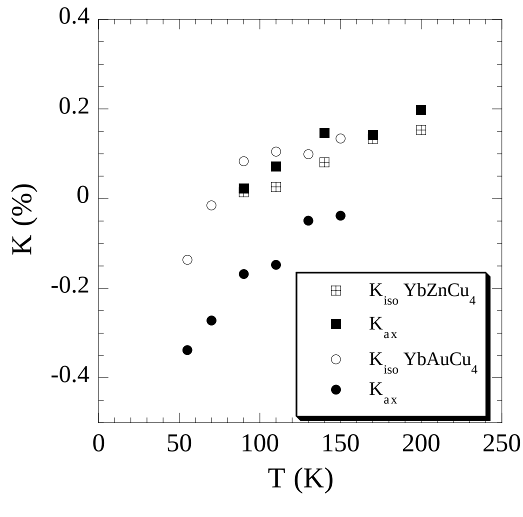

Fig. 1. Temperature dependence of the isotropic and axial knight shift for $YbZnCu_4$ and $YbZnCu_4$.

The ^{63}Cu NMR measurement was carried out under magnetic fields of ~7 T with a phase-coherent pulsed

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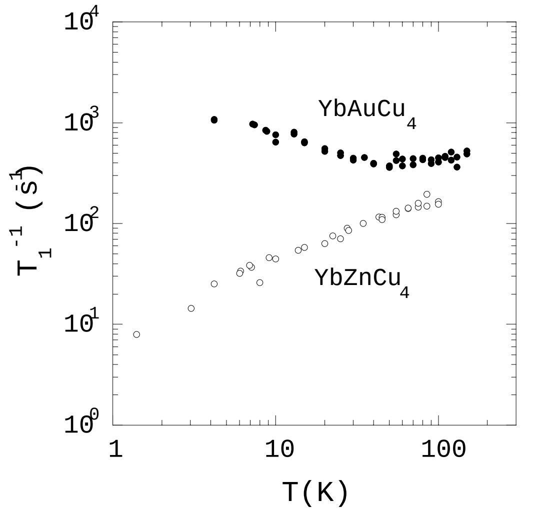

Fig. 2. Temperature dependence of the ^{63}Cu relaxation rate T_1^{-1} in each of $YbZnCu_4$ and $YbAuCu_4$.

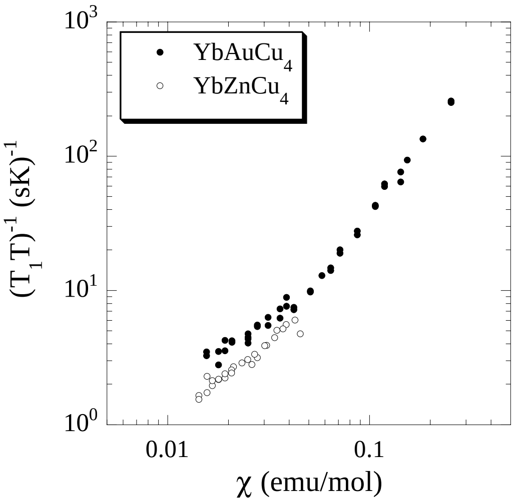

Fig. 3. $(T_1T)^{-1}$ versus χ plots for each of the $YbZnCu_4$ and $YbZnCu_4$ compounds.

spectrometer operating at a contant frequency of 75 MHz. The NMR spectra exhibit the general electric-quadrupole split powder-pattern, and the values of the isotropic Knight shift $K_{\rm iso}$ and axial Kight shift $K_{\rm ax}$ are deduced from the spectrum analysis described in ref. [2]. For the temperatures below $\sim$ 40 K, we could not obtaine any reliable values of the Knight shift, because of the severe broadening of the line width. Fig. 1 shows the temperature dependence of $K_{\rm iso}$ and $K_{\rm ax}$ for $YbZnCu_4$. The data for $YbAuCu_4$ in the figure were cited from our previous report [2]. Both $K_{\rm iso}$ and $K_{\rm ax}$ 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 $H_{\rm hf}^{\rm iso}({\rm tr})$ and $H_{\rm hf}^{\rm ax}({\rm tr})$ as follows: -1.3 and -1.4 kOe/$\mu_{\rm B}$ for $YbZnCu_4$; -0.97 and -1.2 kOe/$\mu_{\rm B}$ 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 *at al.* [3]. The T_1^{-1} data for each of the compounds do not obey the Korringa-like relation (T_1T=const.) within the present experimental temperature range. Then we replotted in Fig. 3 the $(T_1T)^{-1}$ data against the corresponding χ data. For the relaxation process to the fluctuating local moments with the correlation time τ_f, the relaxation rate is given by [4]

$$(T_1T)_f^{-1} = 2z\gamma_{\rm n}^2 k_{\rm B} H_{\rm hf}({\rm tr})^2 \frac{\chi}{\mu_{\rm B} N}\tau_f, \quad (1)$$

assuming a Lorentzian shape of fluctuation spectrum and $\chi(q)\sim\chi(0)$. Here, $\gamma_{\rm n}$ is the nuclear gyromagnetic ratio, z the number of neiboring spins, and N the Avogadro's number. The linear dependence of $(T_1T)^{-1}$ on χ for $YbZnCu_4$ above 1.4 K and for $YbAuCu_4$ above $\sim$50 K indicates that τ_f is nearly independent of the temperature. This is consistent with the strongly localized scheme for the $4f$ moments in these compouns. Taking the experimental values of $(T_1T)^{-1}$ and $H_{\rm hf}^{\rm iso}({\rm tr})$ at high temperatures, we can estimate an order of f spin fluctuation energy T_f $(=\hbar/k_{\rm B}\tau_f)$ 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 χ. 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, τ_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 $T_{\rm K}$. The temperature independent $\tau_f\sim$ for $YbZnCu_4$ down to 1.4 K suggests that the Kondo temperature for $YbZnCu_4$ is much lowere than $T_{\rm K}\sim$30 K estimated from the χ 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.

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