Cu nuclear quadrupole resonance and relaxation on the Yb-based heavy-fermion compound $YbCu_5$

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Cu nuclear quadrupole resonance (NQR) study has been performed on a heavy-fermion compound cubic $YbCu_5$ which has a large electronic specific heat coefficient of $\gamma = 550$ mJ/mol K^2. ^{63}Cu and ^{65}Cu NQR spectra confirmed that magnetic ordering is absent in cubic $YbCu_5$ at least down to 1.6 K. ^{63}Cu nuclear magnetic relaxation rate $(1/T_1)$ of cubic $YbCu_5$ shows Korringa relation where $1/T_1T = \text{const}$ below 4.2 K, indicating the evolution of Fermi-liquid state. With increasing temperature above 50 K, the $4f$-electron contribution to $1/T_1$, $(1/T_1)_{4f}$, tends to be saturated to a constant value, implying that $4f$ electrons are localized with magnetic moments. The temperature dependence of $(1/T_1)_{4f}$ can be qualitatively explained by the temperature dependence of the $4f$-electron correlation rates $(\tau_{4f})^{-1}$, calculated within the impurity Kondo model.
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I. INTRODUCTION

Among many rare-earth intermetallics, the $YbCu_4M$ (M = Ag, Au, Pd, In, Cd, Tl, etc.) compounds with the cubic $AuBe_5$-type structure have been attracting much interest because of their rich variety of low-temperature properties. For example, $YbCu_4In$ exhibits a first-order valence transition from a valence fluctuating state with Pauli-paramagnetic susceptibility to a stable Yb^{3+} state with local magnetic moments at the temperature $T_V = 40$ K with increasing temperature[1,2] or at the magnetic field $H_V = 35$ T with increasing field.[3] On the other hand, $YbCu_4Ag$ shows a typical dense Kondo behavior,[4,5] exhibiting a moderately large electronic specific-heat coefficient of γ being 210–260 mJ/mol K^2 at low temperature.[2,4,6] Furthermore, the temperature dependences of the magnetic susceptibility and the magnetic specific heat are quantitatively explained by the Bethe-ansatz solution of the Coqblin-Schrieffer model with the energy parameter $T_0 = 160$ K.[4,7] As for $YbCu_4Au$ and $YbCu_4Pd$, the existence of long-range magnetic orderings was suggested by the electrical resistivity and the specific-heat measurements.[4] Especially for $YbCu_4Au$, the possibility of incommensurate spin-density wave (SDW) is argued from the result of neutron-diffraction experiment.[8] $YbCu_4Cd$ and $YbCu_4Tl$ are reported to be valence fluctuating compounds.[9] Adding to such varieties of properties, the $YbCu_4M$ system has an advantage of a cubic (the $AuBe_5$-type) structure, which enables us a precise comparison with the theoretical calculations because of its simple crystal structure.

These $YbCu_4M$ compounds have been also studied from a microscopic point of view by many authors in detail. The Cu nuclear quadrupole resonance (NQR) (Ref. 10) and In-NMR (Ref. 11) measurements on $YbCu_4In$ revealed that the temperature dependence of the spin-lattice relaxation rates $1/T_1$ changes drastically from $1/T_1 = \text{const}$ above $T_V = 40$ K to $1/T_1 \propto T$ below T_V, which clearly demonstrates that the $4f$-electron state changes suddenly at $T = T_V$ from a local magnetic moment state to a nonmagnetic Fermi-liquid state with decreasing temperature. The evidence of antiferromagnetic ordering in $YbCu_4Au$ was obtained by the Mössbauer experiment[12] and the Cu-NQR spectrum measurements[13] below 0.7 K. As for $YbCu_4Ag$, the temperature dependence of $1/T_1$ shows the characteristics of a Kondo-lattice system.[13,14] The evaluated value of the characteristic temperature T_0, which is correlated with the Kondo temperature T_K, is in good agreement with that obtained from other measurement.[14]

Recently, we have succeeded in preparing the cubic $AuBe_5$-type $YbCu_5$ (hereafter we describe this compound simply as ''$YbCu_5$'') by high-pressure technique.[15,16] Since $YbCu_5$ might be regarded as the mother compound of all the $YbCu_4M$ systems, the $4f$-electron state of $YbCu_5$ would give significant information about the origin of the various ground states of the $YbCu_4M$ compounds. We have hence investigated the magnetic, electrical, and thermal properties of $YbCu_5$ so far.[16,17] All the results can be consistently interpreted as the characteristics for a Kondo-lattice system with a nonmagnetic Fermi-liquid state below 6 K, i.e., the enhanced Pauli-paramagnetic susceptibility, the T^2 dependence of resistivity, and the T-linear specific heat.[16] Furthermore, the low-temperature state is found to be a heavy-fermion state characterized by the large electronic specific-heat coefficient γ of 550 mJ/mol K^2.[16] It is also notable that $YbCu_5$ shows a metamagnetic-like behavior around $H = 16$ T at low temperature.[16,18]

In this paper, we report on the nuclear quadrupole resonance (NQR) of ^{63,65}Cu in $YbCu_5$ and its spin-lattice relaxation rate $1/T_1$. In order to estimate the contribution from conduction electrons, we measured the $1/T_1$ of Cu NQR in the isostructural $LuCu_4Ag$. We will make a comparison with the theoretical calculation based on the impurity Kondo model.

II. EXPERIMENTAL

The polycrystalline sample of $YbCu_5$ was prepared by high-pressure technique as reported previously.[16] Isostructural $LuCu_4Ag$ was selected as a nonmagnetic reference

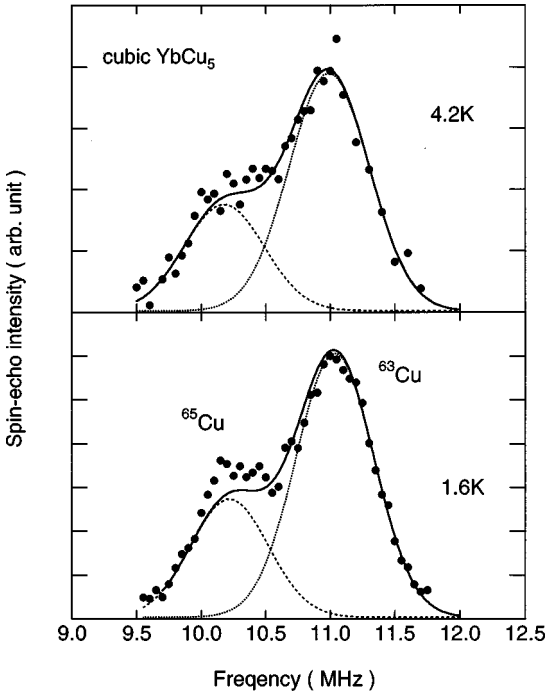

FIG. 1. NQR spectra of $YbCu_5$ at 4.2 and 1.6 K. The solid lines are the results of fitting with a set of two Gaussian functions (see text).

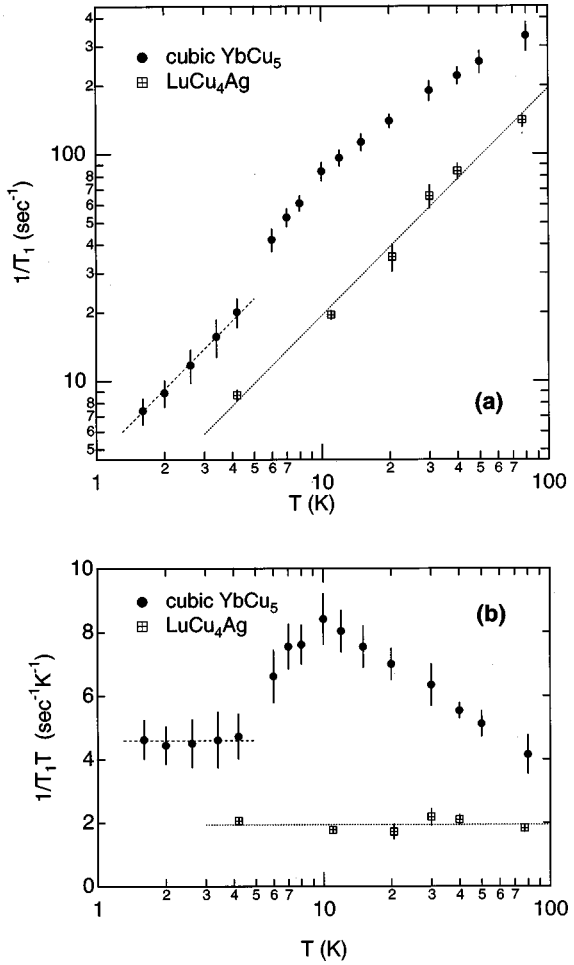

FIG. 2. (a) Temperature dependence of $1/T_1$ for $YbCu_5$ and $LuCu_4Ag$. (b) The temperature dependence of $1/T_1T$. The dotted and broken lines show $1/T_1T=1.9\ \mathrm{sec}^{-1}\mathrm{K}^{-1}$ and $1/T_1T=4.6\ \mathrm{sec}^{-1}\mathrm{K}^{-1}$, respectively.

compound. $LuCu_4Ag$ was prepared by arc melting and subsequent annealing. The sample was crushed into fine powder in order to avoid the eddy-current effect in NQR measurements. Nuclear quadrupole resonance was observed by the conventional spin-echo technique. The spin-lattice relaxation rate $1/T_1$ was measured by the saturation recovery method using the ^{63}Cu-NQR signals. The recovery curve showed single exponential decay over two orders of magnitude. The value of $1/T_1$ was determined by fitting the recovery data to the function expected for the NQR ($\pm\frac{3}{2}>\leftrightarrow\pm\frac{1}{2}>$) transition with the nuclear spin $I=\frac{3}{2}$ as

$$M(t)=M(\infty)[1-\exp(-3t/T_1)], \tag{1}$$

where $M(t)$ is the spin-echo intensity at a long delay time t after the saturation pulse.

III. RESULTS AND DISCUSSION

In Fig. 1 are shown nuclear resonance spectra of $YbCu_5$ under zero magnetic field measured at 1.6 and 4.2 K. These spectra consist of two broad peaks. As is shown in Fig. 1 by the solid lines, we can successfully fit these spectra by a set of two Gaussian functions with the relative intensities being 7:3, equal to the natural abundances of ^{63}Cu and ^{65}Cu, and with the resonance frequencies ν_Q being 10.05 and 11.03 MHz at 1.6 K, of which ratio is equal to that of the nuclear quadrupole moment of ^{63}Cu and ^{65}Cu, $eQ^{63}/eQ^{65}=1.07$. We can therefore assign the spectra to be NQR from ^{63}Cu and ^{65}Cu at crystallographically one site. $YbCu_5$ has two inequivalent Cu sites, i.e., 4*c* and 16*e* sites. The NQR signal is observed when the nucleus is surrounded by the local symmetry lower than cubic. Since the 4*c* Cu site has the cubic local symmetry and the 16*e* Cu site has the lower one than cubic, we can assign these NQR spectra to be that of the 16*e* Cu site. The spectra have similar shape as those of isostructural $YbCu_4Ag$ and nonmagnetic $LuCu_4Ag$, which were already reported.[14] The width of the spectra of $YbCu_5$ are broader than those of $YbCu_4Ag$ and $LuCu_4Ag$ as can be seen from Ref. 14. The width is, however, almost temperature independent between 1.6 and 4.2 K as is seen from Fig. 1. This fact indicates that the width of the spectra is not due to spontaneous or staggered magnetic moment, but is likely due to the local crystalline distortion which is brought during the process of high-pressure synthesizing and/or crushing sample to powders. Thus we can conclude that any kind of magnetic ordering is absent in $YbCu_5$ down to 1.6 K.

In Fig. 2(a) is shown the temperature dependence of the spin-lattice relaxation rate $1/T_1$ of $YbCu_5$ together with that of $LuCu_4Ag$. Figure 2(b) displays plots of $1/T_1T$ vs T. The values of $1/T_1$ of $LuCu_4Ag$ in Ref. 12 are almost identical to $3/T_1$ of our data. This difference arises from the definition of $1/T_1$: we used Eq. (1) while the factor ''3'' in Eq. (1) was omitted in Ref. 12. We measured both $(1/T_1)^{63}$ and $(1/T_1)^{65}$ of $LuCu_4Ag$ at 4.2 K, where $(1/T_1)^{63}$ and $(1/T_1)^{65}$ are $1/T_1$ of ^{63}Cu and ^{65}Cu nuclei, respectively. The ratio $(1/T_1)^{65}/(1/T_1)^{63}$ was observed as 1.162, which is very close to the ratio $(\gamma_N^{65}/\gamma_N^{63})^2=1.147$ where γ_N^{65} and γ_N^{63} are the

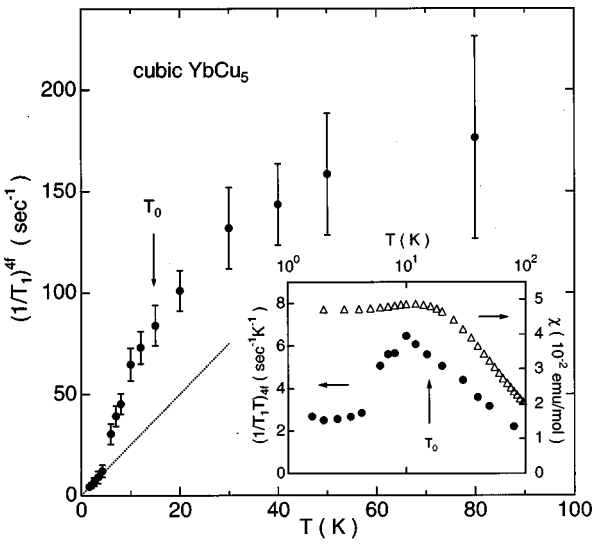

FIG. 3. Temperature dependence of the $4f$-electron contribution to $1/T_1$ of $YbCu_5$, $(1/T_1)_{4f}$. The dotted line indicates the relation $(1/T_1)_{4f} \propto T$. The inset shows the magnetic susceptibility $\chi(T)$ of $YbCu_5$ ($\triangle$) with the low-temperature Curie term subtracted, and $(1/T_1T)_{4f}$ of $YbCu_5$ ($\bullet$). The characteristic temperature $T_0 = 15$ K is indicated by arrow. See text for detail.

gyromagnetic ratio of ^{65}Cu and ^{63}Cu. If the relaxation process is dominated by electric quadrupole effect such as the lattice vibration, the ratio $(1/T_1)^{65}/(1/T_1)^{63}$ should be the value of $(eQ^{65}/eQ^{63})^2 = 0.871$. We can therefore conclude that the relaxation process is magnetic, and the contribution of quadrupole effect to the relaxation can be neglected in this compound. The same situation can be expected for the relaxation of $YbCu_5$. As seen in Fig. 2, $1/T_1$ of $LuCu_4Ag$ is proportional to temperature in the whole temperature range, indicating that the relaxation is dominated by the Korringa-type mechanism. The values of $1/T_1$ of $YbCu_5$ also show the Korringa-type relaxation where $1/T_1 \propto T$ below 4.2 K as is seen in Fig. 2, indicating that the Fermi-liquid state evolves below 4.2 K. This result is consistent with the T^2 dependence of electrical resistivity and the T-linear specific heat, both of which were observed below about 6 K.[16]

In many rare-earth systems, the observed nuclear spin-lattice relaxation rate $(1/T_1)_{\rm obs}$ can be decomposed into two contributions as

$$(1/T_1)_{\rm obs} = (1/T_1)_{4f} + (1/T_1)_s , \tag{2}$$

where $(1/T_1)_{4f}$ and $(1/T_1)_s$ are the contributions due to the fluctuations of Yb-$4f$ moments and the conduction electrons, respectively. We may assume that the values of $(1/T_1)_s$ of $YbCu_5$ can be evaluated by those of $(1/T_1)_{\rm obs}$ of $LuCu_4Ag$. In Fig. 3 are shown the temperature dependence of $(1/T_1)_{4f}$ of $YbCu_5$ estimated by subtracting $(1/T_1)_s = 1.9 \times T\,{\rm sec}^{-1}$ from $(1/T_1)_{\rm obs}$. $(1/T_1)_{4f}$ shows the Korringa-type relaxation below 4.2 K indicating Fermi-liquid state, while it tends to reach a constant value at high temperatures above 50 K. The relation $1/T_1 = {\rm const}$ is a characteristic behavior of the fluctuations due to local magnetic moments.[19] In fact, the magnetic susceptibility of $YbCu_5$ follows the Curie-Weiss behavior with $J = \frac{7}{2}$ and $g_J = \frac{8}{7}$ above 50 K,[16] where J and g_J are the total angular momentum and the Landé factor, respectively. These behaviors are consistent with the Kondo-lattice picture in which a crossover should be observed with increasing temperature from the Fermi-liquid state well below T_K to the local-moment state well above T_K. It should be noted that the magnetic susceptibility of $YbCu_5$ also starts to deviate from the Curie-Weiss behavior with decreasing temperature below about 50 K.[16]

The value of $1/T_1T$ of $LuCu_4Ag$ is estimated to be 1.9 $\sec^{-1}K^{-1}$ from Fig. 2, and that of $YbCu_5$ below 6 K is 4.6 $\sec^{-1}K^{-1}$. So we can evaluate that $(1/T_1T)_s = 1.9\ \sec^{-1}K^{-1}$ whereas $(1/T_1T)_{4f} = 2.7\ \sec^{-1}K^{-1}$. Within the Fermi-liquid description, $1/T_1$ can be related with the density of state (DOS) of the quasiparticle band, $N(E_F)$, as $1/T_1T \propto [A_{hf}N(E_F)]^2$, where A_{hf} is the hyperfine coupling constant between electron and nuclear spins. From the specific heat measurement the DOS of $YbCu_5$ is supposed to be highly enhanced from that of free-electron band.[16] On the other hand, $(1/T_1T)_{4f}$ is the same order of $(1/T_1T)_s$. This fact implies that the value of the transferred hyperfine coupling constant A_{hf} of the ^{63}Cu nuclear spin at 16e site from the $4f$ spin of Yb site is very small, indicating that the hybridization between the Yb $4f$ and the Cu $4s$ or $3d$ of the 16e sites is very weak. We should compare this result with the Cu-NMR study of both the 4c and 16e site, which is now in progress.

It might be interesting to compare the data of $1/T_1$ of $YbCu_5$ with those of typical heavy-fermion compounds $CeCu_6$[20] and $CeRu_2Si_2$,[21,22] since other physical properties of them, especially of $CeRu_2Si_2$, are comparable with those of $YbCu_5$: for instance, large value of γ being 550 mJ/mol K^2 for $YbCu_5$[16] and 350 mJ/mol K^2 for $CeRu_2Si_2$,[23] maxima of susceptibility at $T_{\rm max} = 12$ K for $YbCu_5$,[16] and at $T_{\rm max} = 10$ K for $CeRu_2Si_2$,[21] and metamagnetic behavior in both compounds.[16,23] Therefore we pay attention to the temperature dependence of $1/T_1T$ of $YbCu_5$. As is seen in Fig. 2, $1/T_1T$ of $YbCu_5$ shows a maximum at $T = 10$ K. This behavior is, however, different from those of $CeCu_6$ (Ref. 20) and $CeRu_2Si_2$,[21,22] where $1/T_1T$ shows a constant at low temperature and decreases monotonically above the temperature. On the other hand, very similar behavior to that of $YbCu_5$ is observed for several dense Kondo compounds such as $YbCu_4Ag$ (Ref. 14) and YbCuAl.[24] For these two latter compounds, such temperature dependence of $1/T_1T$ is attributed to the temperature dependence of the $4f$-electron correlation rate $(\tau_{4f})^{-1}$. $(\tau_{4f})^{-1}$ is equal to the quasielastic linewidth of the neutron-scattering energy spectrum Γ. Thus we try to evaluate $(\tau_{4f})^{-1}$ in $YbCu_5$.

$1/T_1$ can be generally expressed with the imaginary part of the dynamical susceptibility as

$$\frac{1}{T_1} = 2\gamma_N^2 k_B T \sum_{\mathbf{q}} [A_{hf}(\mathbf{q})]^2 \frac{\chi''(\mathbf{q},\omega_0)}{\omega_0}, \tag{3}$$

where $A_{hf}(\mathbf{q})$ is the Fourier transform of the hyperfine coupling constant, and ω_0 is the resonance frequency. If we neglect the wave-number dependence and assume the Lorentzian form for the energy spectrum of the $4f$-spin fluctuation with the spectrum width $\Gamma = (\tau_{4f})^{-1}$, we obtain the relation between $(1/T_1)_{4f}$ and $(\tau_{4f})^{-1}$ as

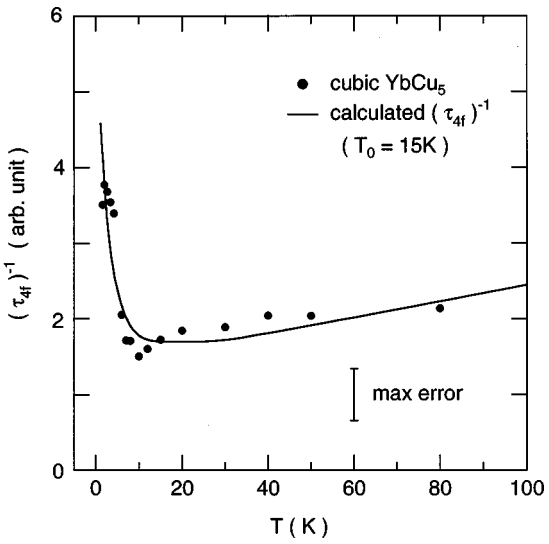

FIG. 4. Temperature dependence of the evaluated $4f$-electron correlation rate $(\tau_{4f})^{-1}$ of $YbCu_5$. The solid line indicates the result of the calculation based on the impurity Kondo model with $T_0=15$ K (see text).

$$\left(\frac{1}{T_1}\right)_{4f}=2\gamma_N^2 k_B A_{hf}^2 T\chi(T)\tau_{4f}, \tag{4}$$

where $\chi(T)$ is the uniform magnetic susceptibility. Therefore $(\tau_{4f})^{-1}$ is proportional to the value of $(T_1)_{4f}T\chi(T)$. The absolute value of $(\tau_{4f})^{-1}$ is, however, unknown since we do not know the value of A_{hf} without any NMR measurement for the moment.

In Fig. 4 is shown the temperature dependence of $(\tau_{4f})^{-1}$ of $YbCu_5$ plotted in arbitrary units. The values of $(\tau_{4f})^{-1}$ show a minimum around 10 K, and continuously increases above the temperature. Such a temperature dependence of $(\tau_{4f})^{-1}$ is often observed for many intermediate valent compounds with nonmagnetic ground state, like $YbInAu_2$ and $CePd_3$,[25] as well as $YbCu_4Ag$ (Ref. 14) and YbCuAl.[24] Cox, Bickers, and Wilkins calculated the temperature dependence of $(\tau_{4f})^{-1}$ based on the impurity Kondo model for a Ce^{3+} $(J=\frac{5}{2})$ ion.[26] Within this model, $(\tau_{4f})^{-1}$ shows a minimum at the temperature around the characteristic temperature T_0. The solid line in Fig. 4 shows the calculation of $(\tau_{4f})^{-1}$ of Cox, Bickers, and Wilkins with $T_0=15$ K. The calculation appears to explain the temperature dependence of $(\tau_{4f})^{-1}$ of $YbCu_5$ qualitatively. It is also notable that both $\chi(T)$ and $(1/T_1T)_{4f}$ of $YbCu_5$ have maxima around T_0 $(=15$ K), as is shown in the inset of Fig. 3. Furthermore, the value of T_0 $=15$ K is in the same order with $T_0=24$ K, which is estimated from the low-temperature specific-heat results.[27] These facts suggest that there exists a single energy parameter T_0 in $YbCu_5$ by which physical properties of $YbCu_5$ are scaled universally.

It must be stressed that the wave-number dependence of the spin fluctuations is assumed to be absent for the calculation of $(\tau_{4f})^{-1}$ by Cox, Bickers, and Wilkins. The relatively good agreement between the calculation and the experimental data may indicate that $YbCu_5$ does not have a strong wave-vector dependence of spin fluctuations even at low temperature. Therefore it can be considered that $YbCu_5$ is the typical heavy-fermion compound where only one parameter T_0 scales the physical properties universally and intersite magnetic correlations is weak, in contrast to those heavy-fermion compounds such as $CeCu_6$ and $CeRu_2Si_2$, where strong antiferromagnetic fluctuations are observed by inelastic neutron scattering at low temperature.[28]

IV. CONCLUSION

We have studied the Cu NQR of the heavy-fermion compound $YbCu_5$. The $^{63}Cu/^{65}Cu$-NQR spectra and the $1/T_1$ of ^{63}Cu have confirmed that $YbCu_5$ does not show any kind of magnetic ordering at least down to 1.6 K. $1/T_1$ shows the Korringa-type relaxation below 4.2 K, while $(1/T_1)_{4f}$ tends to reach a constant value above 50 K. This fact can be considered as the first microscopic evidence of the crossover of $4f$ electrons state in $YbCu_5$ from itinerant Fermi-liquid state below 4.2 K to the stable Yb^{3+} one with local moments well above 50 K. The temperature dependence of $(1/T_1)_{4f}$ can be qualitatively explained by the temperature dependence of the $4f$-electron correlation rate $(\tau_{4f})^{-1}$ and the uniform susceptibility $\chi(T)$. By this comparison between the data and the calculation, the characteristic temperature was estimated as $T_0=15$ K. This value is in good agreement with those estimated from other measurement. The relatively good agreement between the experimental $(\tau_{4f})^{-1}$ and the calculation within the impurity Kondo model may imply that the intersite magnetic correlation is weak in $YbCu_5$ even at low temperature. To discuss the values of $1/T_1$ at high-temperature localized moment state and $1/T_1T$ at Kondo-lattice state at low temperature quantitatively, the absolute values of $(\tau_{4f})^{-1}$ should be determined by NMR measurements, which are now in progress.

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