Anomalous Local Fermi Liquid in $f^2$-Singlet Configuration: Impurity Model for Heavy-Electron System UPt$_3$

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It is shown by the Wilson numerical renormalization group method that a strongly correlated impurity with a crystalline-electric-field singlet groundstate in the $f^2$-configuration exhibits an anomalous local Fermi liquid state in which the static magnetic susceptibility remains an uncorrelated value while the NMR relaxation rate is enhanced in proportion to the square of the mass enhancement factor. Namely, the Korringa-Shiba relation is apparently broken. This feature closely matches the anomalous behaviors observed in UPt$_3$, i.e., the coexistence of an unenhanced value of the Knight shift due to quasiparticles contribution (the decrease across the superconducting transition) and the enhanced relaxation rate of NMR. Such an anomalous Fermi liquid behavior suggests that the Fermi liquid corrections for the susceptibility are highly anisotropic.

1. Introduction

The understanding of heavy electrons of Ce-based compounds with the $f^1$-configuration increased considerably in the 1980s. However, there still remain unresolved issues concerning both normal-state properties and the origin of superconductivity mainly in U-based heavy-electron systems, which are considered to have the $f^2$-configuration, such as UPt$_3$, URu$_2$Si$_2$, and UBe$_{13}$. In particular, it may be one of the milestones that the superconducting states of UPt$_3$ has been identified by Knight shift measurements as the odd-parity state with “equal spin pairing”. By further precise measurements of the $^{195}$Pt Knight shift across the superconducting transition at low magnetic fields and temperatures, it turned out that the Knight shifts of the $b$- and $c$-axes decrease below $T_c$ for $H < 2.3$ kOe. This implies
that the $d$-vector is perpendicular to the $a$-axis. This is consistent with the $E_{1u}$ state, with
\[ d \propto (\hat{k}_a \vec{b} + \hat{k}_b \vec{c})(5\hat{k}_c^2 - 1), \]
in the B-phase (low-$T$ and low-$H$ phase), which was identified by recent thermal conductivity measurements under a rotating magnetic field. \(^8\) However, the decreased amount of Knight shift across the superconducting transition appears as if it is not enhanced by electron correlations and it is nearly the same as that of Pt metal. \(^9\) This implies that the static susceptibility due to the quasiparticles in the sense of the Fermi liquid theory is not enhanced in proportion to the mass enhancement observed in the Sommerfeld constant $\gamma \equiv C/T$. On the other hand, the longitudinal NMR relaxation rate $1/T_1T$ is highly enhanced, \(^10\) reflecting the huge mass enhancement. In this situation, the Korringa-Shiba relation \(^11\) or the conventional Fermi liquid theory is apparently broken. These puzzling properties should be clarified as a first step to elucidate the various characteristic features of UPt$_3$, including the superconducting mechanism.

It has been explained by the slave-boson mean-field approach, on the basis of the extended Anderson model with the $f^2$-crystalline-electric-field (CEF) singlet ground state, \(^12\) that the quasiparticle contribution to the magnetic susceptibility is not enhanced, and it is rather given by that of the hybridization band without correlations. This result is naturally understood if we consider how the magnetic susceptibility due to quasiparticles arises in the case where the local configuration of $f$-electrons is dominated by the $f^2$-CEF singlet as depicted in Fig. 1. Since the $f^2$-CEF singlet state is magnetically inactive except for the Van Vleck contribution, which is incoherent in the terminology of the Fermi liquid theory, the magnetic susceptibility associated with the quasiparticles arises from the $f^1$-states through the hybridization process, $f^2 \to f^1$+conduction electron. Although the $f^1$-state gives the enhanced susceptibility by a factor of $1/z$, its realization probability is given by the renormalization factor $z$. As a result, the static susceptibility exhibits an uncorrelated value. Nevertheless, it still remains unexplained why $1/T_1T$ is enhanced, as in the conventional heavy-electron systems.

The purpose of this paper is to resolve this puzzle at the level of the impurity problem [A short version of this paper has been reported in Ref. 13]. A key point is to understand how the CEF effect in the $f^2$-configuration affects the nature of quasiparticles in the local Fermi liquid. We study the correlated impurity model that possesses essential local correlations as in UPt$_3$ by the Wilson numerical renormalization group (NRG) method. \(^14\)–\(^20\) In particular, we focus on the low-energy renormalized-quasiparticle state under the $f^2$-singlet CEF ground state. In such a case, it has been shown that two types of fixed point exist, i.e., (i) the Kondo-Yosida singlet characterized by the strong-coupling fixed point with the phase shift of $\pi/2$ in both occupied $f$-orbitals, and (ii) the CEF singlet characterized by the phase shift $\delta \approx 0$. \(^18\),\(^21\)
Fig. 1. Schematic picture of quasiparticles, based on the $f^2$-singlet ground state, showing why the quasiparticles give the unenhanced static susceptibility. The quasiparticle susceptibility mainly arises not from the magnetically inactive $f^2$-singlet CEF ground state but from the magnetic $f^1$-state. The weight of the $f^1$-state in the quasiparticles is roughly given by the renormalization factor $z$, which cancels out the enhancement of the susceptibility in the magnetically active $f^1$-state.

When the energy splitting of the CEF is much larger than that of the Kondo temperature $T_K$, the latter case (ii) is realized and the heavy-electron state cannot be formed. In order to form the heavy-electron state in the case of the $f^2$-CEF singlet ground state, it is necessary that the CEF excited states are located close to the singlet ground state because the origin of the heavy mass is the large entropy of local degrees of freedom in general. Then, the density of states of quasiparticles is highly enhanced. The NMR relaxation occurs through the process in which the magnetization of quasiparticles is restored through the flipping of the quasiparticle pseudo-spins with enhanced density of states. This flipping is not suppressed by the renormalization factor $z$ because such an effect has already been taken into account to
suppress the magnetization of quasiparticles. Then, the NMR relaxation rate is expected to be enhanced in proportion to $1/T_K^2$ as in the $f^1$-based heavy-electron compounds.

The organization of this paper is as follows. In Sect. 2, we introduce a pseudo Hund’s rule coupling, which reproduces a low-lying CEF level scheme of the $f^2$-configuration and is incorporated into a two-orbital Anderson model. In Sect. 3, we discuss the physical properties of this model. In Subsects. 3.2 and 3.3, we discuss the relationship between the unenhanced quasiparticle susceptibility and the enhanced NMR relaxation rate. In Subsect. 3.4, we discuss the anisotropy of the magnetic susceptibility due to the CEF effect. In Sect. 4, we discuss the case of the doublet $f^2$-CEF ground state for comparison. Finally, we summarize the results in Sect. 5.

2. Model Hamiltonian

As a minimal model that describes an essential part of local correlations of UPt$_3$, we consider an impurity two-orbital Anderson model with an intra-impurity interaction. The CEF effect in the $f^2$-configuration can be represented by an anisotropic antiferromagnetic Hund’s rule coupling in pseudo-spin space when each Kramers doublet state in the $f^1$-configuration is described by a pseudo-spin, as discussed previously.\textsuperscript{17,18} Here, note that circumstantial evidence for the $(5f)^2$ configuration to be realized in UPt$_3$ was given by high-energy inelastic neutron scattering measurements, which detect the $^3H_4 \rightarrow ^3F_2$ (700 meV) transition in the $f^2$-configuration.\textsuperscript{22}

Now, we recapitulate the previous discussion so as to apply it to UPt$_3$ with the hexagonal symmetry.\textsuperscript{23} Although the CEF ground state of UPt$_3$ is not well identified, it is very suggestive that the temperature dependence of the Knight shift\textsuperscript{6} exhibits behavior similar to that of the static magnetic susceptibility of UPd$_2$Al$_3$,\textsuperscript{24} which also has the same hexagonal symmetry as UPt$_3$. From the analysis of the temperature dependence of the magnetic susceptibility in UPd$_2$Al$_3$,\textsuperscript{24} it was argued that the CEF ground state of the localized $f^2$-component should be the singlet state\textsuperscript{25,26}

$$|\Gamma_4\rangle = \frac{1}{\sqrt{2}}(|+3\rangle - |-3\rangle).$$

From this observation, the ground state of UPt$_3$ is assumed to be $|\Gamma_4\rangle$ in the hexagonal symmetry. To construct this ground state, we take into account two low-lying $f^1$-doublet states ($j_z = \pm 5/2, \pm 1/2$) of three doublet states allowed in the hexagonal CEF in the $j = 5/2$ manifold. The pseudo-spin representation is allotted to the $j_z$-representation of the $f^1$-CEF state
of the impurity as follows:

\[ |+\frac{5}{2}\rangle \equiv |\uparrow, 0\rangle, \quad |-\frac{5}{2}\rangle \equiv |\downarrow, 0\rangle, \quad (2) \]

\[ |+\frac{1}{2}\rangle \equiv |0, \uparrow\rangle, \quad |-\frac{1}{2}\rangle \equiv |0, \downarrow\rangle, \quad (3) \]

where the number 0 indicates that the relevant orbital is unoccupied, e.g., $|\uparrow, 0\rangle$ means that the orbital $|+5/2\rangle$ is occupied and $|\pm 1/2\rangle$ is unoccupied, and $|0, \uparrow\rangle$ means that the orbital $|-1/2\rangle$ is occupied and $|\pm 5/2\rangle$ is unoccupied. The singlet ground state $|\Gamma_4\rangle$ can be represented explicitly in terms of the $f^1$-CEF states in the hexagonal symmetry as follows:

\[ |\Gamma_4\rangle = \frac{1}{\sqrt{2}} \left( |+\frac{5}{2}\rangle|+\frac{1}{2}\rangle - |-\frac{5}{2}\rangle|-\frac{1}{2}\rangle \right) \]

\[ \equiv \frac{1}{\sqrt{2}} \left( |\uparrow, \downarrow\rangle - |\downarrow, \uparrow\rangle \right), \quad (4) \]

where the CEF state in the $f^2$-configuration is formed by the $j$-$j$ coupling scheme and the pseudo-spin state $|\uparrow, \downarrow\rangle$ represents the state such that the orbitals $|+5/2\rangle$ and $|+1/2\rangle$ are occupied. Hereafter, we refer to the channel corresponding to $j_z = \pm 5/2$ as channel 1 and $j_z = \pm 1/2$ as channel 2.

Similarly, the excited CEF states are given as follows:

\[ |\Gamma_3\rangle = \frac{1}{\sqrt{2}} \left( |+3\rangle + |-3\rangle \right) = \frac{1}{\sqrt{2}} \left( |+\frac{5}{2}\rangle|+\frac{1}{2}\rangle + |-\frac{5}{2}\rangle|-\frac{1}{2}\rangle \right) \]

\[ \equiv \frac{1}{\sqrt{2}} \left( |\uparrow, \downarrow\rangle + |\downarrow, \uparrow\rangle \right), \quad (5) \]

\[ |\Gamma^{(2)}_{5+}\rangle = |+2\rangle = |+\frac{5}{2}\rangle|-\frac{1}{2}\rangle \equiv |\uparrow, \uparrow\rangle, \quad (6) \]

\[ |\Gamma^{(2)}_{5-}\rangle = |-2\rangle = |-\frac{5}{2}\rangle|+\frac{1}{2}\rangle \equiv |\downarrow, \downarrow\rangle. \quad (7) \]

Here, (5) is a CEF singlet state, and (6) and (7) are magnetic doublet states in the $f^2$-configuration. The energy separation between (4) and the doublet states (6) and (7) is set to $\Delta$ and that between (4) and (5) is set to $K$, as shown in Fig. 2. Then, the CEF level scheme is reproduced by the effective Hamiltonian

\[ H_{\text{Hund}} = \frac{J_\perp}{2} [S_{1z} S_{2z} + S_{2z} S_{1z}] + J_z S_{1z} S_{2z}, \]

where $\vec{S}_m$ denotes a pseudo-spin operator for a localized electron in orbital $m$, and the couplings $J_\perp$ and $J_z$ are related to $K$ and $\Delta$ as

\[ J_\perp = K \quad \text{and} \quad J_z = 2\Delta - K. \]

\[ \quad (9) \]
Fig. 2. Low-lying CEF energy level scheme of \(f^2\)-configuration in hexagonal symmetry, which is relevant to UPt\(_3\).

Thus, the model Hamiltonian is given by

\[
H = H_K + H_{\text{mix}} + H_f + H_{\text{Hund}},
\]  

with

\[
H_K = \sum_{m=1,2} \sum_{k\sigma} \varepsilon_k c_{km\sigma}^\dagger c_{km\sigma}^\prime,
\]  

\[
H_{\text{mix}} = \sum_{m=1,2} \sum_{k\sigma} (V_{m\sigma} c_{km\sigma}^\dagger f_{m\sigma} + \text{h.c.}),
\]  

\[
H_f = \sum_{m\sigma} E_{f_{m\sigma}} f_{m\sigma}^\dagger f_{m\sigma}^\prime + \sum_{m\sigma} \frac{U}{2} f_{m\sigma}^\dagger f_{m\sigma}^\prime f_{m\sigma}^\dagger f_{m\sigma}^\prime,
\]

where the notations are conventional ones. The term \(H_f\) is introduced to stabilize the \(f^2\)-configuration. Note that the \(k\)-dependence of the hybridization strength is dropped for simplicity hereafter. Although the Hamiltonian [Eq. (10)] is isomorphic with that used in Ref. 18 for discussing the non-Fermi liquid properties observed in Th\(_{1-x}\)U\(_x\)Ru\(_2\)Si\(_2\) (\(x \leq 0.07\)) with tetragonal symmetry, the present CEF level scheme, i.e., a set of \(\Delta\) and \(K\) for UPt\(_3\) with hexagonal symmetry, should be different, and the associated wave functions are also different.

3. Physical properties

In this section, we present the results of NRG calculations\(^{14-16}\) for the model Hamiltonian (8)–(13). We take the unit of energy as \((1 + \Lambda^{-1})D/2\), where \(D\) is half the bandwidth of conduction electrons and \(\Lambda\) is a discretization parameter in the NRG calculation; \(\Lambda = 3\) is used throughout the paper. In each NRG step, up to 600 low lying states are retained. We will use the parameter set, \(E_{f1} = -0.35\), \(E_{f2} = -0.40\), and \(U_1 = U_2 = 1.0\), throughout this paper unless otherwise stated explicitly.
3.1 Kondo effect in the \( f^2 \)-singlet ground state

In this subsection, we first investigate the condition that the huge reduction in the energy scale of local “spin” fluctuations is possible even in the \( f^2 \)-configuration with the CEF singlet ground state. To this end, in Fig. 3, we show a result for the relationship between \( \lim_{\omega \to 0} \Im \chi_\perp(\omega)/\omega \) and \( \Delta \), where \( \Im \chi_\perp(\omega) \) is the imaginary part of the dynamical transverse susceptibility corresponding to the “spin”-flip process between states \( |j_z = +1/2 \rangle \) and \( |j_z = -1/2 \rangle \), and \( \Delta \) is the excitation energy of the doublet state defined above. Explicitly, \( \Im \chi_\perp(\omega) \) at \( T = 0 \) is defined as

\[
\Im \chi_\perp(\omega) = \pi (g \mu_B)^2 \sum_n |\langle n | \hat{j}_+ | 0 \rangle|^2 \times \\
[\delta(\omega - E_n + E_0) - \delta(\omega + E_n - E_0)], \tag{14}
\]

where the Landé factor \( g = 6/7 \), \( \mu_B \) is the Bohr magneton, the transverse component of the one-body total angular momentum operator \( \hat{j}_+ \equiv | +1/2 \rangle \langle -1/2 | \), and \( |n \rangle \) and \( |0 \rangle \) are the excited and ground states of the Hamiltonian (10), with energies \( E_n \) and \( E_0 \), respectively. This definition of \( \Im \chi_\perp(\omega) \) will also be used for that of the NMR relaxation rate, as discussed in Subsect. 3.2. It is remarked here that \( \Im \chi_\perp(\omega) \) is calculated using a standard Kubo formula. The parameters used to calculate the results presented in Fig. 3 are \( V_1 = V_2 = 0.4 \) and \( K = 0.10 \) so as to realize a moderately strong Kondo renormalization, in the case that four CEF states are degenerate, i.e., \( K = \Delta = 0 \). The Kondo temperature \( T_K \) is defined as

\[
1/T_K^2 = \lim_{\omega \to 0} \Im \chi_\perp(\omega)/(g \mu_B)^2 \omega.
\]

The “spin”-flip process, the heart of the Kondo effect, occurs only through the transition between the singlet ground state (4) and the magnetic doublet excited states (6) and (7). In other words, the origin of the large entropy release should be attributed to the existence of the low-lying magnetic doublet states (6) and (7) together with the singlet ground state. As \( \Delta \) becomes larger than a characteristic value on the order of \( T_K^* = \lim_{\omega \to 0} g \mu_B \sqrt{\omega/\Im \chi_\perp(\omega)} \) for \((K, \Delta) = (0.10, 0)\), it becomes difficult for the “spin”-flip process to occur, leading to the suppression of the Kondo effect. Indeed, as \( \Delta \) increases, \( \Im \chi_\perp(\omega)/(g \mu_B)^2 \omega \) decreases monotonically beyond a peak structure around \( \Delta = 0.03 \), which implies that the characteristic energy scale \( T_K \) is greatly suppressed, corresponding to a level crossing of the ground state between the CEF singlet and the Kondo singlet state,\(^{27}\) leading to critical behaviors in many physical quantities. In Refs. 18–20, anomalous non-Fermi liquid properties associated with this criticality have been discussed in detail with an appropriate set of parameters for \( \text{Th}_{1-x} \text{U}_x \text{Ru}_2 \text{Si}_2 \) (\( x \leq 0.07 \)). On the other hand, \( \text{UPt}_3 \) is expected to be located in the Kondo regime, i.e., the left side of the peak in Fig. 3. Therefore,
hereafter, we focus on discussing with the case of $K = 0.10$ and $\Delta = 0.02$, a typical set for the Kondo regime.

![Graph showing the relationship between $\lim_{\omega \to 0} \text{Im} \chi_\perp(\omega)/\omega$ and CEF parameter $\Delta$. A peak structure around $\Delta = 0.03$ arises from the criticality at which the fixed point of this system changes abruptly from the Kondo singlet to the CEF singlet. The unit of $\chi_\perp$ is $(g\mu_B)^2$.

### 3.2 NMR relaxation rate

Next, we discuss the NMR longitudinal relaxation rate $1/T_1$. Longitudinal relaxation occurs through the flipping of the pseudo-spin $j$, or the “real spin” flipping with the orbital angular momentum unchanged.\(^{28}\) In the former case, the relaxation occurs through the pseudo-spin flipping in which the change in $j_z$ is $\Delta j_z = \pm 1$. Since we have discarded the CEF states with $j_z = \pm 3/2$ as irrelevant ones with high excitation energy, the flipping with $\Delta j_z = \pm 1$ occurs only between states with $j_z = \pm 1/2$.

On the other hand, in the latter case, the relaxation is also possible only through the flipping between the states with $j_z = \pm 1/2$ as discussed below. The states of $j_z = \pm 1/2$ are composed of the following states labeled by $l_z$ (orbital angular momentum) and $s_z$ (real spin):

$$
\left| +\frac{1}{2} \right\rangle = \alpha \left| 0, +\frac{1}{2} \right\rangle + \beta \left| 1, -\frac{1}{2} \right\rangle,
\left| -\frac{1}{2} \right\rangle = \alpha \left| 0, -\frac{1}{2} \right\rangle - \beta \left| 1, +\frac{1}{2} \right\rangle,
$$

where $|a, b\rangle$ represents the state of $l_z = a$ and $s_z = b$, and $\alpha$ and $\beta$ are the Clebsch–Gordan coefficients. We have neglected the states with $j_z = \pm 3/2$ because the ground state is assumed
to be $\Gamma_4$, given by Eq. (4). The flipping of the real spin $s_z$ with $l_z$ unchanged is possible only between states $|+1/2\rangle$ and $|-1/2\rangle$. Therefore, in any case, the NMR longitudinal relaxation rate $1/T_1$ is given by $T \lim_{\omega \to 0} \text{Im} \chi_\perp(\omega)/\omega$ with the use of Eq. (14). The result of the NRG calculation of the dependence of $\lim_{\omega \to 0} \text{Im} \chi_\perp(\omega)/(g\mu_B)^2\omega$ on the hybridization $V \equiv V_1 = V_2$ is shown in Fig. 4 for $(K, \Delta) = (0.10, 0.02)$ by filled squares and $(0, 0)$ by filled circles. It is remarked here that $\lim_{\omega \to 0} \text{Im} \chi_\perp(\omega)/(g\mu_B)^2\omega$ for $(K, \Delta) = (0.10, 0.02)$ and that for $(0, 0)$ almost coincide with each other except for when $V \leq 0.5$. For $(K, \Delta) = (0, 0)$, $\lim_{\omega \to 0} \text{Im} \chi_\perp(\omega)/(g\mu_B)^2\omega = 1/T_K^2$ is the definition of the Kondo temperature $T_K$ itself. The open circles are the corresponding quantities for the non-interacting system (without impurity), which are given by $(\chi_{\text{free}}/2)^2$ with $\chi_{\text{free}}$ being the Pauli susceptibility of the non-interacting system defined as

$$\chi_{\text{free}} \equiv \sum_{j_z = 5/2, 1/2} 2(g\mu_Bj_z)^2N(\epsilon_F),$$

where $g = 6/7$. $N(\epsilon_F)$ is the density of states of conduction electrons at the Fermi level for the non-interacting system in each channel with $j_z = 5/2$ or $j_z = 1/2$ and is calculated using the single-particle excitation spectra given by the discretized free-chain Hamiltonian used in the NRG calculations. This result implies that the rate $1/T_1T$ is enhanced in proportion to $1/T_K^2$ as $V$ decreases toward the strongly correlated region. Thus, the NMR relaxation rate seems to directly reflect the effect of the Kondo renormalization. Indeed, for $V = 0.4$, the enhancement in $1/T_1T$ from that for the non-interacting system amounts to about $4 \times 10^2$, implying that the characteristic energy scale $T_K$ is greatly suppressed by about $5 \times 10^{-2}$ to that in the non-interacting limit.

### 3.3 Quasiparticle contribution to susceptibility

Finally, we discuss about the susceptibility from the quasiparticle contribution, which is observed as a decrease in the Knight shift across $T_{c2}$, the lower superconducting transition temperature of UPt$_3$, for a low magnetic field $H < 2.3$ kOe. The decrease in the Knight shift below $T_{c2}$ should be identified as the quasiparticle part of the susceptibility of the system, $\chi_{\text{qp}}$, because the incoherent part due to the Van Vleck contribution should be almost unaffected by the onset of the superconducting state. This separation of the magnetic susceptibility has been established in bulk Fermi liquid theory.\(^{29}\) Namely, the total susceptibility $\chi_z$ consists of two contributions, the incoherent part $\chi_{\text{inc}}$ and the quasiparticle part $\chi_{\text{qp}}$, as shown by Feynman diagrams in Fig. 5. If the magnetization were the conserved quantity, the incoherent contribution $\chi_{\text{inc}}$ would vanish as in the case of the charge susceptibility of single-component
Fig. 4. $1/T_1 T \lim_{\omega \to 0} \text{Im} \chi_{\perp}(\omega)/\omega$ vs $V$. Filled squares represent the case of the CEF-singlet ground state, filled circles represent the quartet state for $K = \Delta = 0$, and open circles represent the non-interacting system. The unit of $\chi_{\perp}$ is $(g\mu_B)^2$.

fermion systems. However, the magnetization in $f$-electron systems with strong spin-orbit interaction is not a conserved quantity so that $\chi_{\text{inc}}$ should remain finite.

The physical picture of quasiparticles in the $f^2$-singlet ground state is shown schematically in Fig. 1. The quasiparticle state can be realized by the process $f^2 \to f^1+$ conduction electron. [In general, $f^3$-states also play similar role to $f^1$-states. However, since the energy levels of $f^3$-states, $E_3^{(1)} (= 2E_{f1} + E_{f2} + U_1) = -0.1$ or $E_3^{(2)} (= E_{f1} + 2E_{f2} + U_2) = -0.15$, in the present model, are moderately higher than those of $f^1$-states, $E_{f1} = -0.35$ or $E_{f2} = -0.40$, the contribution from the $f^3$-states is much less important than that from the $f^1$-states, especially in the low-temperature region. Thus, we neglect the contribution from the $f^3$-states in the quasiparticles.] The $f^2$-singlet ground state (4) has no magnetization. Thus, within the $f^2$-configuration, this ground state can respond only through the Van Vleck term with a virtual transition to the excited CEF state (5). However, such contributions will give the incoherent part $\chi_{\text{inc}}$ of the susceptibility, not the quasiparticle part of the susceptibility $\chi_{\text{qp}}$. In other words, the main contribution to $\chi_{\text{qp}}$ arises from the part of the $f^1$-states in quasiparticles, and that from the $f^3$-states is negligible, as mentioned above. From these considerations, it is reasonable to define the quasiparticle susceptibility $\chi_{\text{qp}}$ as the difference between $\chi_z$ and $\chi_{\text{inc}}$:

$$\chi_{\text{qp}} = \chi_z - \chi_{\text{inc}}, \quad (18)$$

where $\chi_z$ is the total static susceptibility including the contribution of all $f^n$-states ($n =$
and $\chi_{\text{inc}}$ is the incoherent part of the susceptibility, which consists of the contributions from $f^n$-states with $n = 0$ and $2 - 4$. It is almost evident that $f^n$-states with $n = 0$ and $2 - 4$ contribute to $\chi_{\text{inc}}$.

However, the separation of $\chi_{\text{qp}}$ and the part of $\chi_{\text{inc}}$ arising from $f^1$-states is nontrivial. Indeed, an incoherent contribution $\chi_{\text{inc}}$ also exists in the case where the $f^1$-configuration is dominant, as in Ce-based heavy-fermion systems, if the Van Vleck contribution, arising from the virtual transition between the ground and excited CEF states in the $f^1$-configuration, exists. Such a condition is satisfied in the case where the magnetization operator $\hat{m}_z = \mu_B(\hat{l}_z + 2\hat{s}_z)$ has off-diagonal matrix elements between the ground and excited CEF states, e.g., $\Gamma_7^0 (\sqrt{1/6}|\pm 5/2\rangle - \sqrt{5/6}|\mp 3/2\rangle)$ and $\Gamma_8^0 (\sqrt{5/6}|\pm 5/2\rangle + \sqrt{1/6}|\mp 3/2\rangle)$ states in the cubic symmetry. However, in the hexagonal system, as in the present case, where $f^1$-CEF states are given by the eigenstates of $\hat{j}_z, |\pm 1/2\rangle, |\pm 3/2\rangle$, and $|\pm 5/2\rangle$, $\hat{m}_z$ has no off-diagonal matrix elements among these states. Therefore, it is considered that the contribution from $f^1$-states to $\chi_{\text{inc}}$ can be safely neglected, justifying the definition of the quasiparticles contribution $\chi_{\text{qp}}$ [Eq. (18)] to the magnetic susceptibility.

In the NRG calculation, the magnetic susceptibility is calculated using the linear response of the magnetic moment to the tiny magnetic field $H$, as $\chi = \langle m \rangle / H$, where $\langle m \rangle = g\mu_B\langle \sum_{j_z=\pm 1/2,\pm 3/2} \hat{f}_j^\dagger \hat{f}_j \rangle$ is the average of magnetic moment in the ground state. In practical calculation, the magnetic moment for states with different numbers of $f$ electrons can be calculated separately. For example, $m = g\mu_B j_z$, with $g = 6/7$, for the $f^1$-state $\hat{f}_j^\dagger |0\rangle$, $m = g\mu_B (|\pm 1/2\pm 5/2\rangle)$ for the $f^2$-state $\hat{f}_{\pm 1/2}^\dagger \hat{f}_{\pm 5/2}^\dagger |0\rangle$, and so on. Thus, we can separately analyze the contributions to the susceptibility from the states with different numbers of $f$-electrons.

![Fig. 5. Structure of Feynman diagrams giving the magnetic susceptibility. In general, the magnetic susceptibility is separated into the quasiparticle contribution (in the dashed box) and the incoherent one, $\chi_{\text{inc}}$. The quasiparticle susceptibility can be described by the propagator of the quasiparticle (solid lines with arrow), the Fermi liquid correction (squares), and the effective magnetic moment (triangles).]
First, we discuss the relationship between the static susceptibility $\chi_z$ of the impurity and the energy separation $K$ between the two CEF singlets. The result is shown in Fig. 6 for a typical case with $\Delta = 0.02$. The dashed curve represents the $f^2$-contribution, which we denote as $\chi_{\text{inc}}$ implying the incoherent contribution. For comparison, the Van Vleck contribution $\chi_{\text{vv}}$ arising from the two singlets of the localized orbital [given by Eqs. (4) and (5)] is represented by the dotted line. Explicitly, $\chi_{\text{vv}}$ is given by

$$\chi_{\text{vv}} = \frac{\langle \Gamma_3 | J_{\text{inc}}^{\text{at}} | \Gamma_4 \rangle^2}{E_3 - E_4} = \frac{(3g\mu_B)^2}{K},$$

and does not include the many-body effect with conduction electrons. Note that as $K$ increases, $\chi_{\text{inc}}$ decreases monotonically. On the other hand, $\chi_{\text{qp}} = \chi_z - \chi_{\text{inc}}$ shows almost no change with $K$.

![Fig. 6](image)  

**Fig. 6.** $K$ dependence of the total static magnetic susceptibility (solid line), the incoherent part of susceptibility (dashed line), and the Van Vleck contribution $\chi_{\text{vv}}$ from the isolated local degrees of freedom with $V = 0$ (dotted line). The unit of $\chi_z$ is $\mu_B^2$.

Next, in Fig. 7, we show the relationship between $\chi_{\text{qp}}$ at $T = 0$ for $K = 0.10$ and $\Delta = 0.02$ and the hybridization $V$, and compare it with the Pauli susceptibility $\chi_{\text{free}}$ for the non-interacting system (without an impurity) defined by Eq. (17). Note that the order of magnitude of $\chi_{\text{qp}}$ is the same as that of $\chi_{\text{free}}$ for a wide range of $V$. This is compatible with the fact that the decrease in the Knight shift across $T_{c2}$ observed in UPt$_3$ is nearly the same as the Knight shift of Pt metal,\(^9\) and is consistent with the theoretical result obtained by the slave-boson mean-field treatment for the lattice version of the present model.\(^{12}\) The values of $\chi_{\text{qp}}$ for
$V \geq 0.5$ even agree quantitatively with those of $\chi_{\text{free}}$. The quantitative discrepancy for $V \leq 0.5$ may be attributed to the residual interaction among local quasiparticles, a part of the Fermi liquid interaction. However, it should be noted that such an enhancement in $\chi_{\text{qp}}$ compared with $\chi_{\text{free}}$ is only a factor of 2 for $V = 0.4$, and is much smaller than 20, which corresponds to the enhancement in $1/T_1T$ for the same hybridization parameter as shown in Subsect. 3.2. It is a nontrivial situation that the quasiparticle contribution is not enhanced, while the characteristic energy scale $T_K$ is suppressed considerably as in the heavy electron systems.

Thus, the so-called Korringa-Shiba relation is apparently broken in this situation.

### 3.4 Anisotropy of the CEF effect in the $f^2$-configuration

We have shown in previous subsections that an anomalous local Fermi liquid appears due to the CEF effect on the renormalized quasiparticles in the $f^2$-configuration, and that the singlet CEF ground state plays an essential role for almost unenhanced longitudinal quasiparticle susceptibility. As a next step, it is important to know which state, magnetic doublet states or a singlet state, is appropriate for the first excited state to explain the behavior of UPt$_3$. We have assumed $K > \Delta$ in Subsects. 3.2 and 3.3. Here, we investigate the origin of the difference in the renormalization effect between the longitudinal and transverse susceptibilities by comparing the behavior of magnetic susceptibilities for $K > \Delta$ with that for $K < \Delta$.

First, we analyze the CEF effect on the dynamical structure of two responses under the
We show the $\omega$-dependence of the imaginary part of the dynamical susceptibilities, $\text{Im}\chi_z(\omega)$ for the longitudinal response and $\text{Im}\chi_{\perp}(\omega)$ for transverse response, for $K > \Delta$ ($K, \Delta$)=(0.1, 0.02) in Fig. 8 and $K < \Delta$ ($K, \Delta$)=(0.01, 0.05) in Fig. 9.

![Graph](image)

**Fig. 8.** (Color online) CEF effect on the spectral weight of transverse and longitudinal susceptibilities for $K > \Delta$, $K = 0.1$, and $\Delta = 0.02$. The parameters are $E_{f1} = E_{f2} = -0.40$, $U_1 = U_2 = 1.0$, and $V_1 = V_2 = 0.4$. The values of $\text{Im}\chi_z$ are normalized by those for $S = 1/2$.

To obtain isotropic responses for $K = \Delta = 0$, two $f^1$-levels are set to be the same, $E_{f1} = E_{f2} = -0.40$, and the absolute value of $\text{Im}\chi_z(\omega)$ is normalized by that for $S = 1/2$. Without the CEF effect, for $(K, \Delta)=(0, 0)$, the two responses (triangles and inverse triangles) coincide with each other as expected in both figures. By the CEF effect, for $(K, \Delta)=(0.1, 0.02)$ [$K > \Delta$] in Fig. 8, the peak of $\text{Im}\chi_z(\omega)$ (closed circles) is suppressed and is shifted to the high energy side corresponding to the excitations of the Van Vleck term. On the other hand, that of $\text{Im}\chi_{\perp}(\omega)$ (closed squares) is enhanced and shifted to the low-energy side corresponding to the reduction in the characteristic energy scale $T_K$ of magnetic fluctuations. On the other hand, for $(K, \Delta)=(0.01, 0.05)$ [$K > \Delta$] in Fig. 9, the tendencies of the two responses are interchanged. Namely, the peak of $\text{Im}\chi_z(\omega)$ is enhanced while the values of $\text{Im}\chi_{\perp}(\omega)$ are suppressed as a whole. Note that, in both cases, a new pronounced magnetic excitation appears at a lower energy than the peak position for $(K, \Delta) = (0, 0)$ in the isotropic case. This indicates that the entropy release of $f$-electrons due to the Kondo effect is prevented by the presence of the CEF splitting, but the existence of the spectral weight at lower energy implies that the Kondo effect eventually occurs. This implies that the entropy of $f$-electrons still remains in a low-
energy region because the Kondo effect still survives against the tendency of forming a local CEF singlet.

Secondly, to clearly understand the relationship between \((K, \Delta)\) and \((\chi_z, \text{Im}\chi_\perp)\), we show in Fig. 10 the \(K\)-dependence of the two responses \(\chi_z/(g\mu_B)^2\) at \(T=0\) and \(\lim_{\omega \to 0}[\text{Im}\chi_\perp(\omega)/2\pi\omega]^{1/2}/(g\mu_B)\) together with the Sommerfeld constant \(\gamma\), which is calculated by the numerical differentiation of entropy with respect to \(T\). Here, \(\chi_z\) is also normalized by that of \(S=1/2\) so as to fulfill the Korringa-Shiba relation\(^{11}\) for \(K=\Delta=0\):

\[
\lim_{\omega \to 0} \frac{\text{Im}\chi_\perp(\omega)}{2\pi\omega} = \frac{\chi_z^2}{(g\mu_B)^2}.
\]

The Sommerfeld constant \(\gamma\) increases with increasing \(K\), so that the effective mass is enhanced and \(T_K\) is suppressed by the CEF effect, as discussed above. In the case of \(K<\Delta\), the value of the longitudinal susceptibility \((\chi_z)\) is larger than that of the transverse susceptibility \((\chi_\perp)\). As \(K\) increases, \(\chi_z\) decreases, while \(\chi_\perp\) is enhanced in proportion to \(\gamma\). This result shows that \(\chi_\perp\) directly reflects the effect of mass enhancement while \(\chi_z\) does not for \(K>\Delta\). This indicates that the situation of \(K>\Delta\) may be realized to be consistent with the observed behavior of UPt\(_3\), i.e., the unenhanced Knight shift and the enhanced relaxation rate of NMR.\(^{7,9}\)
Fig. 10. Two components of susceptibility $\chi_z/(g\mu_B)^2$ at $T = 0$ and $\lim_{\omega \to 0} \left[ \text{Im} \chi_\perp(\omega)/2\pi\omega \right]^{1/2}/(g\mu_B)$ together with the Sommerfeld constant $\gamma$ as a function of $K$. Each component is divided by the appropriate coefficient to have the same dimension.

Fig. 11. Longitudinal and transverse susceptibilities $\chi_z/(g\mu_B)^2$ at $T = 0$ and $\lim_{\omega \to 0} \left[ \text{Im} \chi_\perp(\omega)/2\pi\omega \right]^{1/2}/(g\mu_B)$ together with the Sommerfeld constant $\gamma$ as a function of $K$. Each component is appropriately scaled. For $\Delta < 0$, the ground state is doublet.

4. Case of Doublet $f^2$-CEF Ground State

In this section, we investigate the nature of quasiparticles in the case of the $f^2$-doublet ground state. We show in Fig. 11 the same physical quantities as shown in Fig. 10 when
\( \Delta \) is varied with fixed \( K \). As \( \Delta \) decreases, for \( \Delta < 0 \), the CEF ground state changes from the singlet to the magnetic doublet state. In the case of the \( f^2 \)-doublet ground state, \( \chi_z \) is enhanced in proportion to \( \gamma \), reflecting the effect of mass enhancement. This is incompatible with the observed behavior of \( \text{UPT}_3 \). \( \chi_{\perp} \) is almost independent of \( \Delta \) in the range shown in this figure. This is because, when \( |\Delta| \) increases, the suppression of the characteristic energy scale is almost canceled out by the decrease in the probability of the transition between the CEF singlet state and the doublet states.

5. Conclusion

We have shown by NRG calculations that the impurity with the \( f^2 \)-CEF ground state can behave as an anomalous local Fermi liquid, which is characterized by unrenormalized quasiparticle longitudinal susceptibility and enhanced transverse susceptibility, reflecting the suppression of energy scales of magnetic fluctuations. This gives a crucial hint to understand the peculiar NMR behaviors observed in \( \text{UPt}_3 \).

In the heavy-electron state with the \( f^2 \)-singlet CEF ground state, the NMR relaxation rate is enhanced in proportion to \( 1/T_k^2 \) because it has the same origin as the Kondo effect, i.e., the “spin”-flip process, while the quasiparticle susceptibility \( \chi_{\text{qp}} \) is not enhanced by a correlation effect. The static susceptibility is dominated by the Van Vleck contribution arising from virtual excitation processes among CEF levels in the \( f^2 \)-configuration, while the quasiparticle susceptibility \( \chi_{\text{qp}} \), arising from one-electron excitations, remains the same order of magnitude as that of a non-interacting system. Owing to such highly anisotropic behaviors in the magnetic susceptibilities, the Korringa-Shiba relation is broken. Such a situation is realized when the first excited CEF state in the \( f^2 \)-configuration is a magnetic doublet state, which has matrix elements with the singlet ground state through the “spin”-flip process. This result confirms the physical picture obtained previously by the slave-boson mean-field approach for a lattice system.

In Sect. 4, we have discussed the difference in the renormalization effect on magnetic susceptibilities for other CEF schemes in the \( f^2 \)-configuration. We concluded that such an unenhanced quasiparticle susceptibility is realized only in the case of the \( f^2 \)-singlet ground state. When the ground state is a magnetic doublet state, the static susceptibility is enhanced, reflecting the effect of the mass enhancement.

We have confirmed that such anomalous behaviors are obtained for a wide range of parameter sets simulating \( \text{UPT}_3 \) with the hexagonal symmetry. However, the highly anisotropic magnetic property of local quasiparticles in \( f^2 \)-based systems is ubiquitous. Namely, they
are expected to also be realized for other systems with a nonmagnetic singlet CEF ground state, regardless of the system being located in the region of either the Kondo or CEF singlet state. For example, a cubic or tetragonal system with a singlet CEF ground state in the $f^2$-configuration is also expected to exhibit magnetic properties similar to UPt$_3$. On the other hand, in a cubic system, the critical behaviors due to the transition between two singlet states, Kondo and CEF, may be different from that in the case of Th$_{1-x}$U$_x$Ru$_2$Si$_2$ ($x \leq 0.07$) with the tetragonal symmetry\cite{18}, as suggested by the results reported in Refs. 31 and 32. Thus, the highly anisotropic Fermi liquid is expected to be a natural consequence of heavy-fermion systems with the $f^2$ CEF singlet configuration.

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