Non-Fermi Liquid Behavior of U-impurity with $f^2$-Singlet Ground State

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It is shown by the Wilson’s numerical renormalization-group approach that Non-Fermi liquid (NFL) behaviors arise even for U-impurity with an $f^2$-singlet ground state, and all the puzzling behaviors of $R_{1-x}U_xRu_2Si_2$ ($R=\text{Th, Y and La}$, $x \leq 0.07$) can be explained on its basis. There exists an unstable NFL fixed point due to the balance between Kondo singlet and Crystalline-Electric-Field singlet ground states. For wide region in parameter space the system shows the NFL behavior as a transient phenomenon in temperature range accessible by experiment. Results for temperature dependence of the resistivity, the magnetic susceptibility and the Sommerfeld coefficient with and without magnetic field reproduce the behaviors observed in $\text{Th}_{1-x}U_xRu_2Si_2$. Less anomalous properties of $R_{1-x}U_xRu_2Si_2$ ($R=\text{Y and La}$) can be understood along the same scenario provided that those compounds have different sets of parameters.

KEYWORDS: Non-Fermi liquid, numerical renormalization-group method, Crystalline-Electric-Field singlet, $R_{1-x}U_xRu_2Si_2$ ($R=\text{Th, Y and La}$)

Recently, issues about the Non-Fermi liquid (NFL) behaviors in dilute U arrays have attracted much attention. These are related to a general interest of Kondo phenomena which arise in some cases due to the existence of plural number of strongly correlated $5f$ electrons per U ion. Especially, the behaviors in series of compounds $R_{1-x}U_xRu_2Si_2$ ($R=\text{Th, Y and La}$, $x \leq 0.07$) have been well investigated as a single-site effect, because anomalous properties of these materials are well scaled by impurity concentrations. The latter fact makes the theoretical approach simple compared with the system in which the intersite effect is considered to play some role, such as $\text{Y}_{1-x}U_x \text{Pd}_3$. $\text{Th}_{1-x}U_xRu_2Si_2$ shows $-\ln T$ divergence both of the susceptibility $\chi$ and the Sommerfeld coefficient $\gamma$ with decreasing temperature. However, a big puzzle still remains as discussed shortly.

The logarithmic dependence of these thermodynamic quantities itself is consistent with the prediction based on the quadrupolar Kondo model if it is generalized appropriately. The quadrupolar Kondo effect is expected to arise for a non-Kramers doublet ground state of $f^2$-configuration and to be mapped onto the $S=1/2$ two-channel Kondo model. The exact solution of $S=1/2$ two-channel Kondo model predicts the asymptotic behavior of $\chi$, $\chi \propto -\ln T$ and the resistivity $\rho \propto \mathrm{const} + \sqrt{T}$ in the limit $T \rightarrow 0$ with the residual entropy $S(T \rightarrow 0) = \frac{1}{2} \ln 2$. Since $R_{1-x}U_xRu_2Si_2$ ($R=\text{Th, Y and La}$) has the tetragonal symmetry and the valence of the uranium ion is expected to be mainly $U^{4+}$ ($5f^2$), many theoretical studies of this problem have been done on the basis of the two-channel Kondo model assuming the magnetic doublet ground state, $|T_5^{(2)}\pm\rangle$, in ($5f^2$) configuration.

However, there exist puzzling properties about $\text{Th}_{1-x}U_xRu_2Si_2$ which cannot be explained by the two-channel Kondo model. First of all, $\chi$ shows the Curie law while $\rho$ shows large value of the order of the unitarity limit for $10K < T < 100K$. Namely, the susceptibility indicates the existence of the localized moment while the resistivity suggests the confinement of the localized moment by the spin polarization of conduction electrons. This apparent inconsistency was removed by the extended two-channel Anderson model introduced and discussed by Sakai and his collaborators who showed that there exists wide set of parameters which gives results consistent with the experiments of $\text{Th}_{1-x}U_xRu_2Si_2$. However, in the case of $R_{1-x}U_xRu_2Si_2$ ($R=\text{Y and La}$), $\rho$ is almost constant below 10K, which cannot be solved by such an extended two-channel Anderson model. Secondly, the divergence of $\gamma$ is suppressed for applied magnetic field $H$ of the order of several Tesla in $\text{Th}_{1-x}U_xRu_2Si_2$. In any theories based on the two-channel Kondo model, the entropy is expected to be released at finite temperature because the magnetic field lifts the degeneracy of the $|T_5^{(2)}\pm\rangle$ doublet ground state, leading enhancement of $\gamma$. Thus, the theory based on the two-channel Kondo model with magnetic doublet configuration does not seem to be a final theory for this problem.

The purpose of this paper is to solve this puzzle on the basis of a novel mechanism assuming Crystalline-Electric-Field (CEF) singlet ground state, in which the competition between the conventional Kondo singlet and CEF singlet gives rise to the NFL behavior in low but intermediate temperature regime as a transient phenomenon. A system exhibiting such competition can be mapped to the $f^2$-impurity model with anisotropic antiferromagnetic Hund’s-rule coupling and different hybridizations with conduction electrons, when CEF levels are specified by pseudo spins. It has been recog-
nized by recent studies that such models exhibit the NFL behavior reminiscent of two-channel Kondo model for certain critical parameter sets. On the basis of Wilson’s numerical renormalization-group (NRG) method, we show that the NFL behaviors arise, as a transient phenomenon near the unstable fixed point, rather robustly even for parameters off the criticality in rather wide temperature region accessible by experiments. Series of present results are consistent with those observed in R_{1−x}Ru_{x}Si_{2} (R=Th, Y and La). Especially, all the anomalous properties of Th_{1−x}Ru_{x}Si_{2} can be explained semiquantitatively for a certain set of parameters.

We consider two low-lying $f^{1}$ doublet states out of three doublet of $j = 5/2$ orbitals in the tetragonal symmetry. These four (two doublet) states can be represented by pseudo spin:

$$\left| \Gamma^{(2)}_{1}, + \right\rangle = -\sqrt{\frac{1}{6}} \left| \frac{5}{2} \right\rangle + \sqrt{\frac{5}{6}} \left| \frac{3}{2} \right\rangle \equiv | \uparrow, 0 \rangle,$$

$$\left| \Gamma^{(2)}_{1}, - \right\rangle = \sqrt{\frac{1}{6}} \left| \frac{5}{2} \right\rangle - \sqrt{\frac{5}{6}} \left| \frac{3}{2} \right\rangle \equiv | \downarrow, 0 \rangle,$$

$$\left| \Gamma_{2}, + \right\rangle = \frac{1}{2} \left| 0, \uparrow \right\rangle,$$

$$\left| \Gamma_{2}, - \right\rangle = -\frac{1}{2} \left| 0, \downarrow \right\rangle,$$

where $| \uparrow, 0 \rangle$ ($| 0, \uparrow \rangle$) represents the state that the pseudo spin in channel 1 (2) is up and the channel 2 (1) is empty, and notations are conventional otherwise. In terms of these four states, the relevant CEF states in $f^{2}$-configuration are written in the $j$-$j$ coupling scheme as follows:

$$\left| \Gamma_{4} \right\rangle = \frac{1}{\sqrt{2}} (| 2 \rangle - | -2 \rangle) = \frac{1}{\sqrt{2}} (| \uparrow, \downarrow \rangle - | \downarrow, \uparrow \rangle),$$

$$\left| \Gamma_{3} \right\rangle = \frac{1}{\sqrt{2}} (| 2 \rangle + | -2 \rangle) = \frac{1}{\sqrt{2}} (| \uparrow, \downarrow \rangle + | \downarrow, \uparrow \rangle),$$

$$\left| \Gamma^{(2)}_{5}, + \right\rangle = \beta | 3 \rangle - \alpha | -1 \rangle = | \uparrow, \uparrow \rangle,$$

$$\left| \Gamma^{(2)}_{5}, - \right\rangle = \beta | -3 \rangle - \alpha | 1 \rangle = | \downarrow, \downarrow \rangle.$$  

Here, the energy levels of $| \uparrow, 0 \rangle$ and $| 0, \uparrow \rangle$ have been omitted because of large intra-orbital Coulomb repulsion. One of fundamental assumptions is to adopt CEF level scheme shown in Fig.1. This level scheme can be reproduced by the pseudo-spin Hamiltonian

$$H_{\text{Hand}} = \frac{J_{1}}{2} \left[ S_{1}^{z} S_{2}^{z} + S_{1}^{z} S_{2}^{z} \right] + J_{2} S_{1}^{z} S_{2}^{z},$$

where $S_{m}$ denotes a pseudo-spin operator of the localized electrons in the orbital $m$. By taking such the pseudo spin representation, it can be understood clearly that the CEF singlet competes with the Kondo singlet directly and we can perform calculations more accurately owing to an appearance of new conserved quantity, pseudo spin. The energy separation between the singlet ($\Gamma_{4}$) and the doublet ($\Gamma^{(2)}_{5}$)± is set to be $K$ and that between the $\Gamma_{4}$ and the singlet ($\Gamma_{3}$) is set to be $\delta$. The couplings $J_{1}$ and $J_{2}$ are related to $(K, \delta)$ as $J_{1}=K$ and $J_{2}=2\Delta - K$.

It is remarked that (0.9) represents an anisotropic antiferromagnetic Hund’s-rule coupling, which cannot exist in real spin system.

With this preliminary, the system in question can be described by a conventional two-channel Anderson model with antiferromagnetic Hund’s-rule coupling:

$$H = H_{K} + H_{\text{mix}} + H_{f} + H_{\text{Hand}},$$

with

$$H_{K} \equiv \sum_{m=1,2} \sum_{\kappa \sigma} \epsilon_{\kappa} c_{\kappa m \sigma}^\dagger c_{\kappa m \sigma},$$

$$H_{\text{mix}} \equiv \sum_{m=1,2} \sum_{\kappa \sigma} (V_{\kappa, m} c_{\kappa m \sigma}^\dagger f_{m \sigma} + \text{h.c.}),$$

$$H_{f} = \sum_{m \sigma} E_{m} f_{m \sigma}^\dagger f_{m \sigma} + \sum_{m \sigma} \frac{U_{m}}{2} f_{m \sigma}^\dagger f_{m \sigma} f_{m \sigma} f_{m \sigma}.$$
In summary, we have shown that the puzzling problem of Th$_{1-x}$Ru$_2$Si$_2$ ($x \leq 0.07$) can be fully understood on the basis of a mechanism characterized by Non-Fermi liquid like transient phenomenon with $f^2$-singlet ground state. A key assumption is that the system corresponding to Th$_{1-x}$Ru$_2$Si$_2$ is incidentally located near the critical points. However, it is not so artificial to assume such an incident if one considers the fact that the NFL behaviors appear weakly in much more restricted temperature range for R$_{1-x}$U$_x$Ru$_2$Si$_2$ ($R =$ Y and La) which would have different set of parameters due to the difference of conduction band structure. Experimental results show that $T_{cr}$ in the case of $R =$ La is about 10K while that in the case of $R =$ Th has not been observed. This indicates that the parameter set of La$_{1-x}$U$_x$Ru$_2$Si$_2$ would be located less close to the critical points than that of Th$_{1-x}$U$_x$Ru$_2$Si$_2$. If $T_{cr}$’s are increased with unchanged CEF level structure, the final fixed point can be changed from the CEF singlet to the Kondo singlet. Experimentally, when high pressure is applied, these materials may show the increase of the resistivity with decreasing temperature corresponding to the change of the fixed point.

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Fig. 1. CEF level scheme in $f^2$-configuration adopted in this paper.

Fig. 2. Phase diagram of the ground state in $K$-$\Delta$ plane. Squares represent the critical points determined by NRG calculations. The cross represents a parameter set used in this paper.

Fig. 3. The renormalization flow below $T < T_{K1}$. The NFL behavior should be observed when the system passes near the critical point.

Fig. 4. Temperature dependence of the magnetic susceptibility $\chi(T)$ (triangles) and frequency dependence of the scattering rate $\tau^{-1}(\omega)$ at $T = 0$ (squares).

Fig. 5. Contribution of each channels $\tau^{-1}(\omega)$ (circles) and $\tau^{-1,2}(\omega)$ (triangles) to the total scattering rate $\tau^{-1}(\omega)$ (squares).

Fig. 6. Temperature dependence of the Sommerfeld coefficient $\gamma \equiv C/T$ under the magnetic field of $H = 10^{-8}$ (squares) and $H = 10^{-3}$ (circles). The inset shows temperature dependence of the entropy. The other parameter set is the same as shown in Fig.4 and Fig.5.