Enhanced Valence Fluctuations Caused by f-c Coulomb Interaction in Ce-Based Heavy Electrons:
Possible Origin of Pressure-Induced Enhancement of Superconducting Transition Temperature in CeCu$_2$Ge$_2$ and Related Compounds

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Properties of an extended periodic Anderson model with the f-c Coulomb interaction $U_{fc}$ is studied as a model for CeCu$_2$Ge$_2$ and related compounds which is considered to exhibit a sharp valence change under pressure. The problem is treated by extending the *slave-boson* and large-$N$ expansion method to treat the present system. It is shown that, as the f-level $\epsilon_f$ is increased relative to the Fermi level, the sharp valence change is caused by the effect of $U_{fc}$ with moderate strength of the order of the bandwidth of conduction electrons. The superconducting transition temperature $T_c$ due to the valence-fluctuation exchange is estimated on the slave-boson fluctuation approximation. In the model with spherical Fermi surface, $T_c$ exhibits sharp peak as a function of $\epsilon_f$, simulating the effect of pressure, for the $d$-wave pairing channel.

KEYWORDS: CeCu$_2$Ge$_2$, f-c Coulomb interaction, valence transition, unconventional superconductivity

§1. Introduction

CeCu$_2$Si$_2$ is well known not only as the firstly discovered unconventional superconductor with $T_c \sim 0.7$ K at ambient pressure,\textsuperscript{1)} but as a remarkable pressure dependence of $T_c$ exhibiting a pronounced peak at $P \simeq 17$ GPa.\textsuperscript{2)} The isostructural compound, CeCu$_2$Ge$_2$, has a similar phase diagram in $P$-$T$ plane in which one can see the superconductivity appears after the magnetism is suppressed by pressure at $P \sim 8$ GPa.\textsuperscript{3–5)} These two compounds have similar physical properties, if the scale of the pressure for CeCu$_2$Si$_2$ is shifted by 7.6 GPa.\textsuperscript{6)} The mechanism of such prominent enhancement of $T_c$ has not been understood so far. A purpose of this paper is to discuss it from the viewpoint that such phenomena is based on the sharp valence change of Ce ion.

Apart from the pronounced peak of $T_c$, remarkable properties under pressure of these compounds are as follows:\textsuperscript{3)} 1) The residual resistivity $\rho_0$ also exhibits a peak at around the pressure where

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$T_c$ takes the maximum. 2) The coefficient $A$ of the $T^2$-term of resistivity rapidly decreases by about two orders of magnitude at around the same pressure as $T_c$ and $\rho_0$ exhibit peaks. These phenomena may be attributed to a rapid change of the valence of Ce ion,\(^7\) while the origin of the superconductivity around magnetic quantum critical point (QCP) is considered to be induced by the enhanced spin fluctuations.\(^8\) It is not only because the position of the sharp peak of $T_c$ is located far away from the pressure corresponding to QCP, but the valence of Ce ion seems to decrease rapidly there. The latter fact is supported by the rapid decrease of the coefficient $A$ by about $10^{-3}$ times, which implies that the Sommerfeld constant $\gamma$ decreases by more than $10^{-1}$ times suggesting that the system is changed from the Kondo regime to the valence fluctuation (VF) regime rapidly. This is also supplemented by the rapid decrease of the so-called Kadowaki-Woods ratio $A/\gamma^2$ by 25 times there from the value of heavy electrons to that of the conventional transition metals.\(^4, 9-11\)

Recently, we have proposed that the physics behind these intriguing behaviors should be the enhanced valence fluctuations of Ce ion, and given a consistent explanation on the basis of a phenomenological model for the valence fluctuations.\(^7\) In the present paper we develop its microscopic theory on the basis of an extended periodic Anderson model with repulsion $U_{fc}$ between f- and conduction electrons. Without $U_{fc}$, the valence change does not occur so drastically. On the other hand, it is shown on the mean-field approximation of slave bosons that the rapid valence change can be realized for moderate strength of $U_{fc}$ of the order of the bandwidth of conduction electrons if the level of f-electron $\epsilon_f$ is tuned relative to the Fermi level. It is also shown by taking into account the fluctuations of slave bosons beyond the mean-field values that the superconductivity can be induced in the d-wave channel associated with the rapid valence change provided that the spherical conduction band is adopted.

In §2, we introduce the extended periodic Anderson model and briefly review the relevant works performed so far. In §3, we present a formalism for obtaining mean-field solutions in the slave-boson approach and taking into account the fluctuations around it. The results of mean-field solutions of slave bosons and superconducting transition temperature are presented in §4. The main results of this paper are as follows: 1) Sharp valence change is caused by the effect of $U_{fc}$ with moderate strength of the order of the bandwidth of conduction electrons, when the f-level $\epsilon_f$ is tuned as a mimic of the effect of the pressure. 2) The superconducting state is induced by the process of exchanging the slave-boson fluctuations for the values of $\epsilon_f$ at which the sharp valence change occurs. 3) The symmetry of so induced superconducting state is d-wave if the spherical model is adopted for conduction electrons. The discussions are given in §5.

§2. Extended Periodic Anderson Model with f-c Coulomb Interaction

The effects of pressure appear through changes of the parameters characterizing the physics, the Kondo coupling $J$, the hybridization matrix element $V$, and the f-level $\epsilon_f$. However, these variations of parameters themselves cannot afford causing the drastic change of valence of Ce-ion
from that of the Kondo regime to the VF regime, which leads to the divergent increase of the valence susceptibility, while the valence increases prominently as the strength of $V$ is increased or the level $\epsilon_f$ is increased approaching the Fermi level.\(^\text{12}\) Here we take into account the effect of the short range Coulomb repulsion between f electron and conduction electrons, because the repulsion may promote the charge transfer between both type of electronic states in general when it is combined with the level shift of $\epsilon_f$. Namely, we extend the conventional periodic Anderson model (PAM) as follows:

$$
H = \sum_{k\sigma}(\epsilon_k - \mu)c_{k\sigma}^\dagger c_{k\sigma} + \epsilon_f \sum_{k\sigma} f_{k\sigma}^\dagger f_{k\sigma} + U_{ff} \sum_i n_{i\uparrow}^f n_{i\downarrow}^f + V \sum_{k\sigma}(c_{k\sigma}^\dagger f_{k\sigma} + \text{h.c.}) + U_{fc} \sum_{i\sigma\sigma'} n_{i\sigma}^f n_{i\sigma'}^c,
$$

where the conventional notations for PAM are used except for $U_{fc}$, the f-c Coulomb repulsion.

The effect of $U_{fc}$ on the valence of Ce-ion has been discussed in a number of contexts and models. First, with regard to the impurity Anderson model, the effect of $U_{fc}$ was discussed in relation to optical experiments, valence-band photoemission (PES) and bremsstrahlung isochromat spectroscopy (BIS).\(^\text{13–16}\) Costi and Hewson studied the effects of $U_{fc}$ by a numerical renormalization group (NRG) approach.\(^\text{15,16}\) They concluded that in the Kondo regimes $U_{fc}$ can be absorbed to other parameters ($V$, $\epsilon_f$ and $U_{ff}$), and the same set of renormalized parameters on $U_{fc} = 0$ is consistent with both the valence-band PES and BIS spectra and the thermodynamic properties. The effect of increasing $U_{fc}$ is to increase the Kondo temperature, thus to decrease the number of f-electrons $n_f$ per ion mored rapidly. Indeed, a related model has been investigated on the basis of NRG approach by Takayama and Sakai, and it turned out that $n_f$ rapidly decreases at $\epsilon_f \approx E_F - U_{fc}$, $E_F$ being the Fermi level, as $\epsilon_f$ is increased so as to approach $E_F$.\(^\text{17}\) This result implies that the rapid valence change occurs where $\epsilon_f + U_{fc}$, the energy of $f^1$ state, and $E_F$, the energy of the state $f^0$+(extra conduction electron on the Fermi level), are nearly degenerate giving rise to enhanced valence fluctuations. Then, $U_{fc}$ has a considerable effect on the valence fluctuation, although it does not cause a valence instability in the impurity Anderson model.

On the other hand, it is not easy to examine the effects of $U_{fc}$ in the PAM due to the lattice effect. In the case of impurity model, the conduction band plays a role of the electron bath so that the chemical potential $\mu$ is essentially fixed. However, in the case of lattice model, $\mu$ is considerably affected by $U_{fc}$ itself so that we have to treat the problem in a self-consistent fashion. In particular, such a self-consistent treatment is indispensable when the valence state of Ce begins to leave from the Kondo regime to VF one under the high pressure. There exist some studies of the extended PAM with $U_{fc}$ within Hartree-Fock like approximations.\(^\text{18–20}\) Even in this simple level of approximation, $U_{fc}$ is responsible for the rapid change of the number of f-electrons as the level $\epsilon_f$ is tuned, leading to the first order transition in the large $U_{fc}$ region. Recently, we have investigated the extended PAM with $U_{fc}$ by the variational Monte Carlo method on the extended Gutzwiller variational wave
It has been shown there that the valence of f-electrons decreases rapidly as the level of f-electrons $\epsilon_f$ is increased, if $U_{fc}$ is moderately large comparable to half of the bandwidth of conduction electrons.

The similar effects of the d-p Coulomb interaction in the so-called d-p model have been investigated as a possible charge fluctuation mechanism of the high $T_c$ superconductor. On the other hand, the physics discussed in the present paper is rather different from “valence fluctuation mechanism” proposed for heavy electron superconductors in Ref. 24.

On the basis of these considerations, we investigate the effect of $U_{fc}$ in the extended PAM as a possible origin of the salient phenomena observed in CeCu$_2$Ge$_2$ under high pressures, particularly a possible mechanism of the enhancement of the superconducting transition temperature which has not been clarified so far. We study the extended PAM with $U_{fc}$ by the slave-boson and large-N expansion approach, which has been shown to be effective for studying thermodynamics properties, superconducting transition temperature, transport properties, etc. of the usual PAM. Following these methods, we extend it so as to include the f-c Coulomb interaction within a Gaussian fluctuation approximation and study its effects on the superconducting transition temperature.

§3. Formulation

In this section we summarize a formulation for calculations using the slave-boson large-N expansion technique. We generalize the Hamiltonian eq. (2.1) to the case where the N-fold degeneracy exists, including both spin and orbital degrees of freedom. Then, we introduce the slave-boson operators to exclude the double occupied state by f-electrons assuming $U_{ff} = \infty$. Thus, the Hamiltonian is expressed as follows:

$$\mathcal{H} = \sum_{k m} \left( \epsilon_k c_k^\dagger c_k + \epsilon_f f_k^\dagger f_k \right) + \frac{V}{\sqrt{N}} \sum_{k q m} \left( c_{k m}^\dagger c_{k+q m} b_q^\dagger + \text{h.c.} \right) + U_{fc} \sum_{i m} f_i^\dagger f_i c_i^\dagger c_i, \quad (3.1)$$

where $b$'s are slave-boson operators describing f$^0$ state and the constraint

$$Q_i = \sum_m f_i^\dagger f_i + b_i^\dagger b_i = 1 \quad (3.2)$$

is required at each site in order to maintain an equivalence of the truncated Hilbert space and the original one. To generate a 1/N-expansion we rewrite variables as follows:

$$\begin{align*}
Q_i & \rightarrow \frac{Q_i}{N} \\
b & \rightarrow \frac{b}{\sqrt{N}} \\
V & \rightarrow \frac{V}{\sqrt{N}} \\
U_{fc} & \rightarrow \frac{U_{fc}}{\sqrt{N}}
\end{align*} \quad (3.3)$$

Hereafter, we use the radial gauge following Ref. 25. (Although the radial and the Cartesian gauge formulation are ultimately equivalent, spurious infrared divergences do not appear in the
In the above expressions, we have rewritten variables $f$ and $\lambda'$ as $f'$ and $\lambda$ respectively, and neglected the Jacobian factor $\prod_{i\tau} \rho_i(\tau)$ following Ref. 25.

By introducing two kinds of Stratonovich-Hubbard fields $\varphi^f$ and $\varphi^c$ for $U_{fc}$, we can perform the functional integral over the Fermion fields in eq. (3.4) and eq. (3.5). Then, the partition function $Z$ is given by

$$Z = \int D(\rho \lambda \varphi^f \varphi^c) \exp(-S),$$

(3.6)

where

$$S = \int_0^\beta d\tau \left[ \sum_{kk'} f'_{km}(\tau) \{(\partial_{\tau} + \epsilon_k) \delta_{kk'} + \frac{1}{\sqrt{N_L}} i\lambda(k^\prime - k; \tau)\} f'_{k'm}(\tau) \right. \left. + \sum_{km} c'_{km}(\tau) \{(\partial_{\tau} + \epsilon_k) c_{km}(\tau) + \frac{V}{\sqrt{N_L}} \sum_{kk'} \{ c_{km}(\tau) f_{k'm}(\tau) \rho(k^\prime - k; \tau) + \text{h.c.} \} \right. \left. + i\frac{N}{\sqrt{N_L}} \sum_{kk'} \rho(-k^\prime; \tau) \lambda(k^\prime - k; \tau) \rho(k; \tau) - i\eta_0 N \sqrt{N_L} \lambda(0; \tau) + \frac{U_{fc}}{N} \sum_{ilm} n_{il}^f n_{im}^c \right].$$

(3.5)

In the above expressions, we have rewritten variables $f'$ and $\lambda'$ as $f$ and $\lambda$, respectively, and performed a local gauge transformation, $b_i(\tau) = \rho_i(\tau) e^{i\theta_i(\tau)}$, $f_{im}(\tau) = f'_{im}(\tau) e^{i\theta_i(\tau)}$ and $\lambda'_i(\tau) \equiv \lambda_i + \dot{\theta}_i(\tau)$. Then, the partition function is given by

$$Z = \int D(\rho \lambda \varphi^f \varphi^c) \exp(-S),$$

(3.4)

$$S = -N \text{Tr} \ln \hat{A} + i \frac{N}{\sqrt{N_L}} T^2 \sum_{kk'} \rho(-k) \lambda(k^\prime - k) \rho(k)$$

$$- i\eta_0 N \sqrt{N_L} \int d\tau \lambda(0; \tau) - \frac{NU_{fc}}{4} T \sum_k \varphi^f(k) \varphi^c(-k),$$

(3.7)

where we have introduced abbreviation $k = (k, \omega_n)$, etc. and matrix $\hat{A}$ is defined as

$$A_{kk'} = \begin{bmatrix} 0 & -i\omega_n + \epsilon_k & (U_{fc}/2\sqrt{N_L}) \varphi^f(k - k') & T \sqrt{N_L} \rho(k - k') \\ -i\omega_n - \epsilon_k & 0 & T \sqrt{N_L} \rho(k - k') & \varphi^f(k - k') \\ T \sqrt{N_L} \rho^*(k - k') & \varphi^c(k - k') & -i\omega_n + \epsilon_k & T \sqrt{N_L} \rho(k - k') \\
T \sqrt{N_L} \rho^*(k - k') & \varphi^c(k - k') & 0 & -i\omega_n - \epsilon_k + \frac{U_{fc}}{2\sqrt{N_L}} \varphi^f(k - k') + \frac{T \sqrt{N_L} \rho(k - k')}{T \sqrt{N_L} \rho^*(k - k')} \end{bmatrix}.$$

(3.8)

At the level of mean-field approximation, the action $S_0$ can be written as

$$S_0 = -N \text{Tr} \ln \hat{A}_0 + \frac{iN}{T} \frac{\beta^2}{\sqrt{N_L}} - q_0 \sqrt{N_L} \bar{\lambda} - \frac{NU_{fc}}{4T} \varphi^f(k) \varphi^c(-k)$$

$$= -N \sum_k \ln \left[ (i\omega_n - \epsilon_k - \frac{U_{fc}}{2\sqrt{N_L}} \varphi^f) (i\omega_n - \epsilon_k - \frac{i\bar{\lambda}}{\sqrt{N_L}} - \frac{U_{fc}}{2\sqrt{N_L}} \varphi^f) - \frac{V^2}{N_L} \beta^2 \right]$$

$$+ N \frac{i}{T} \left( \frac{\beta^2}{\sqrt{N_L}} - q_0 \sqrt{N_L} \bar{\lambda} - \frac{NU_{fc}}{4T} \varphi^f \varphi^c, \right.$$

(3.9)
where we have approximated $\lambda$, $\rho$ and $\varphi$ by their uniform mean field value as

$$
\begin{align*}
\lambda(q) &= \frac{1}{T} \bar{\lambda} \delta_q \\
\rho(q) &= \frac{1}{T} \bar{\rho} \delta_q \\
\varphi(q) &= \frac{1}{T} \bar{\varphi} \delta_q
\end{align*}
$$

and

$$
A_{0 \, kk'} = \begin{bmatrix} -i \omega_n + \epsilon_k & \frac{V}{\sqrt{N L}} \bar{\rho} \nu f \\
\frac{V}{\sqrt{N L}} \bar{\rho} & -i \omega_n + \epsilon_f + \frac{i \bar{\lambda}}{\sqrt{N L}} + \frac{U_{fc}}{2 \sqrt{N L}} \bar{\varphi} \end{bmatrix} \delta_{kk'}
$$

$$= -\hat{G}_0^{-1}. \tag{3.12}
$$

Here $\hat{G}_0$ is nothing but a matrix of Green’s functions for the renormalized band. In this mean-field approximation, quasiparticles acquire the heavy effective mass through the strong f-f Coulomb interactions, $U_{ff} = \infty$, and deep f-level $\epsilon_f$ far below the Fermi level. From the optimum conditions for mean fields, $\partial S_0 / \partial \bar{\rho} = 0$, $\partial S_0 / \partial \bar{\lambda} = 0$ and $\partial S_0 / \partial \bar{\varphi} = 0$, we obtain the following set of self-consistent equations:

$$
i \bar{\lambda} \sqrt{N L} = -\frac{T}{N L} \sum_k \frac{V^2}{(i \omega_n - \epsilon_k)(i \omega - \epsilon_f) - V^2}, \tag{3.13a}
$$

$$q_0 - \left( \frac{\bar{\rho}}{\sqrt{N L}} \right)^2 = \frac{T}{N L} \sum_k \frac{i \omega_n - \bar{\epsilon}_k}{(i \omega_n - \epsilon_k)(i \omega - \epsilon_f) - V^2}, \tag{3.13b}
$$

$$\frac{\bar{\varphi}^f}{2 \sqrt{N L}} = \frac{T}{N L} \sum_k \frac{i \omega_n - \epsilon_k}{(i \omega_n - \epsilon_k)(i \omega - \epsilon_f) - V^2}, \tag{3.13c}
$$

$$\frac{\bar{\varphi}^c}{2 \sqrt{N L}} = \frac{T}{N L} \sum_k \frac{i \omega_n - \epsilon_f}{(i \omega_n - \tilde{\epsilon}_f)(i \omega - \epsilon_f) - V^2}, \tag{3.13d}
$$

where we have introduced abbreviations as

$$
\tilde{\epsilon}_k \equiv \epsilon_k + U_{fc} \tilde{n}_f, \tag{3.14a}
$$

$$\tilde{\epsilon}_f \equiv \epsilon_f + i \lambda / \sqrt{N L} + U_{fc} \tilde{n}_c, \tag{3.14b}
$$

$$\tilde{V} \equiv V \bar{\rho} / \sqrt{N L}. \tag{3.14c}
$$

where $\tilde{n}_f \equiv \langle n_f \rangle_{MF}$, the f-electron numbers per site and “spin”, and $\tilde{n}_c \equiv \langle n_c \rangle_{MF}$, the number of conduction electrons per site and “spin”. It is noted that both the left-hand sides of (3.13b) and (3.13c) are equal to $\tilde{n}_f$ and that of (3.13d) is equal to $\tilde{n}_c$. Thus, the following relations hold:

$$
q_0 - \left( \frac{\bar{\rho}}{\sqrt{N L}} \right)^2 = \frac{\bar{\varphi}^f}{2 \sqrt{N L}} = \tilde{n}_f \tag{3.15}
$$

$$\frac{\bar{\varphi}^c}{2 \sqrt{N L}} = \tilde{n}_c. \tag{3.16}
$$

In terms of these quantities, the matrix Green function $\hat{G}_0$, eq. (3.12), can be expressed as

$$
\hat{G}_0 = \frac{1}{(i \omega_n - \epsilon_k)(i \omega - \epsilon_f) - V^2} \begin{bmatrix} i \omega - \epsilon_f & \tilde{V} \\
\tilde{V} & i \omega - \tilde{\epsilon}_k \end{bmatrix}. \tag{3.17}
$$
The first term of eq. (3.21) can be expanded as

We truncate this expansion at \( n = 1 \) obtaining a Gaussian form. Then, we can write the partition function of the Gaussian fluctuation part as (See Appendix B)

\[
S = S_0 + \tilde{S},
\]

where \( S_0 \) is given by (3.10) and the fluctuation part \( \tilde{S} \) is given as follows:

\[
\tilde{S} = -N \text{Tr} \ln \tilde{A} + \frac{iN}{\sqrt{N_L}} T \sum_k \{ \tilde{\lambda} \tilde{\rho}(-k) \tilde{\rho}(k) + 2 \tilde{\rho} \tilde{\lambda}(-k) \tilde{\rho}(k) \} - \frac{NUc}{4} T \sum_k \tilde{\varphi}^t(k) \tilde{\varphi}(k)
\]

Then, we expand \( S, (3.7) \), with respect to the fluctuations up to the second order as

\[
S = S_0 + \tilde{S},
\]

where \( S_0 \) is given by (3.10) and the fluctuation part \( \tilde{S} \) is given as follows:

\[
\tilde{S} = -N \text{Tr} \ln \tilde{A} = -N \text{Tr} \ln(1 - \tilde{G}_0 \tilde{M}) = \sum_{n=1}^\infty \text{Tr}(\tilde{G}_0 \tilde{M})^n.
\]

We truncate this expansion at \( n = 2 \) obtaining a Gaussian form. Then, we can write the partition function of the Gaussian fluctuation part as (See Appendix B)

\[
\tilde{Z} = \int D(\tilde{\rho} \tilde{\lambda} \tilde{\varphi} \tilde{\varphi}^t) \exp[-\tilde{S}],
\]

\[
\tilde{S} = NT \sum_k (\tilde{\rho}(-k), i\tilde{\lambda}(-k), \tilde{\varphi}(-k), \tilde{\varphi}^t(-k)) \tilde{S}_k (\tilde{\rho}(k), i\tilde{\lambda}(k), \tilde{\varphi}(k), \tilde{\varphi}^t(k))^t,
\]

where the superscript \( t \) means the transpose is taken, and the symmetric matrix \( \tilde{S}_k \) is given as

\[
\tilde{S}_k = \begin{bmatrix}
S_{\rho\rho}(k) & S_{\rho\lambda}(k) & S_{\rho\varphi}^t(k) & S_{\rho\varphi^t}(k) \\
S_{\lambda\rho}(k) & S_{\lambda\lambda}(k) & S_{\lambda\varphi}^t(k) & S_{\lambda\varphi^t}(k) \\
S_{\varphi^t\varphi}(k) & S_{\varphi^t\varphi}(k) & S_{\varphi^t\varphi^t}(k) & S_{\varphi^t\varphi^t}(k) \\
S_{\varphi^t\varphi^t}(k) & S_{\varphi^t\varphi^t}(k) & S_{\varphi^t\varphi^t}(k) & S_{\varphi^t\varphi^t}(k)
\end{bmatrix},
\]

\[
\tilde{S}_k = \begin{bmatrix}
\frac{i\tilde{\lambda}}{\sqrt{N_L}} + V^2 \Pi^t(k) + V^2 \Pi_3(k) & \frac{\tilde{\rho}}{\sqrt{N_L}} + V \Pi_2(k) & \frac{Uc}{2} \Pi \Pi_2(k) & \frac{Uc}{2} \Pi \Pi_1(k) \\
\frac{1}{2} \Pi^t(k) & \frac{1}{2} \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k) \\
\frac{1}{2} \Pi^t(k) & \frac{1}{2} \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k) \\
\frac{1}{2} \Pi^t(k) & \frac{1}{2} \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k) & \frac{Uc}{2} \Pi \Pi^t(k)
\end{bmatrix}.
\]
where the definition of polarization functions $\Pi$’s are given in Appendix B, and the symmetric off-diagonal components have abbreviated. The inverse matrix of $\hat{S}_k$ gives the matrix Green Functions for bose fields

$$D_{\alpha\beta}(k; \tau) \equiv -\langle T^\tau \alpha(k, \tau) \beta^\dagger(k, 0) \rangle,$$

(3.28) where $\alpha$, $\beta$ are $\tilde{\rho}$, $\tilde{\lambda}$, $\tilde{\varphi}^f$, $\tilde{\varphi}^c$, as

$$\hat{D}(k) = -\hat{S}^{-1}(k).$$

(3.29)

In Fig. 1, we show the various possible interaction terms included in eq. (3.21) by Feynman diagrams.

Fig. 1. Feynman diagrams of interaction terms of eq. (3.21). (a) represents the hybridization process. (c), (d) and (g) are new terms which appear through the $f$-$c$ Coulomb interaction $U_{fc}$. (e) and (f) represent the second term of eq. (3.21), and (g) corresponds to the last term.

§4. Results

4.1 Mean-Field Solutions

First, we present the solutions within the mean-field approximation. Self-consistent equations (3.13) can be rearranged as follows:\(^{(31)}\)

$$\frac{i\tilde{\lambda}}{\sqrt{N_l}} = \frac{V^2}{N_L} \sum_k \frac{f(E_k^-) - f(E_k^+)}{\sqrt{\left(\tilde{\varepsilon}_i - \tilde{\varepsilon}_k\right)^2 + 4V^2}}$$

(4.1a)

$$q_0 - \left(\frac{\tilde{\rho}}{\sqrt{N_L}}\right)^2 = \frac{1}{2N_L} \sum_{k, \pm} \left[1 \pm \frac{\tilde{\varepsilon}_i - \tilde{\varepsilon}_k}{\sqrt{(\tilde{\varepsilon}_i - \tilde{\varepsilon}_k)^2 + 4V^2}}\right] f(E_k^\pm)$$

(4.1b)

$$\frac{1}{N_L} \sum_k \{f(E_k^-) + f(E_k^+)\} = \tilde{n}_f + \tilde{n}_c$$

(4.1c)

where $f(x)$ is the Fermi distribution function and $E^\pm_k$ are the quasi particle energies

$$E_k^\pm = \frac{1}{2} [\tilde{\varepsilon}_i + \tilde{\varepsilon}_k \pm \sqrt{(\tilde{\varepsilon}_i - \tilde{\varepsilon}_k)^2 + 4V^2}].$$

(4.2)
A set of equations (4.1) is equivalent to that for the case of conventional PAM without $U_{fc}$ except for the point that $\bar{\epsilon}_k$ and $\bar{\epsilon}_f$ include the term arising from $U_{fc}$ as seen in (3.14). We solve them for a three-dimensional model with the dispersion $\epsilon_k$ for the conduction-electrons approximated by that of free electron, $\epsilon_k = k^2/2m - D$, where the bottom of the conduction band is set as $-D$. Hereafter we use $D$ as the unit of energy. The bare mass, $m$, of conduction electrons is chosen such that the integration of, $\rho_0(\epsilon)$, the density of state per “spin” of conduction electrons with respect to $\epsilon$ from $-D$ to $D$ is equal to 1:

$$\rho_0(\epsilon) = \frac{3}{4\sqrt{2}D} \sqrt{\frac{\epsilon + D}{D}}.$$  (4.3)

In Fig. 2, we show the results of $\bar{n}_f$ as a function of $\epsilon_f$ for several values of $U_{fc}/D$. Here we set the hybridization as $V = 0.5D$ and total electron number per “spin” as $n = \bar{n}_f + \bar{n}_c = 0.875$. These results are consistent with those of previous works.\textsuperscript{18–20} These results are consistent with a physical picture that the rapid valence change occurs at $\epsilon_f \approx E_F - U_{fc}$ where the energies of $f^0$- and $f^1$-state are degenerate leading to enhanced valence fluctuations. For much larger values of $U_{fc}$ or smaller values of $V$ than those presented in Fig. 2, there occurs a first-order like discontinuous transitions although they are not shown in Fig. 2. In the mean-field level of approximation for slave boson Hamiltonian, our treatment of the effect of $U_{fc}$ is just like that in the Hartree-Fock approximation.\textsuperscript{18–20} It is noted that the valence change occurs more sharply if we estimate it in much more proper approximation on the extended Gutzwiller variational wave function.\textsuperscript{21} In this sense, the sharpness of the valence change may be underestimated by the present treatment.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{$\bar{n}_f$, f-electron number per site and “spin”, as a function of $\epsilon_f$, f-electron level, in three-dimensinal model where the free electron dispersion $\epsilon_k = k^2/2m - D$ is adopted for the conduction electrons.}
\end{figure}

Using the mean-field solution we calculate the density of states $\rho(\mu)$ of quasiparticles at the Fermi
energy. It is related to the Sommerfeld constant $\gamma$ of the specific heat as

$$\gamma = \frac{\pi^2}{3} D \rho(\mu) N,$$  \hspace{1cm} (4.4)

where $N$ is the degeneracy of the quasiparticles. The so-called Kondo temperature, or the characteristic temperature, of the present model, is defined simply as $(\bar{\epsilon} - \mu)$. In Fig. 3, we can see that the relation $T_K \propto \gamma^{-1}$ holds. It is remarked that the relation $T_K$ vs. $\gamma$ with different value of $U_{fc}$ are lying on a line exhibiting a kind of universality. It is suggested that the effect of $U_{fc}$ can be absorbed into the other parameters of the conventional PAM at least in the mean-field level, like in the case of the single impurity Anderson model. However, in the lattice case, the first-order like transition occurs through the effects of $U_{fc}$ even in the mean-field approximation. This is considered to be a distinctive effect of $U_{fc}$ in the PAM.

Fig. 3. Sommerfeld constant $\gamma$ vs Kondo temperature $T_K$, defined by $T_K = \bar{\epsilon} - \mu$. The parameters used are the same as those in Fig. 2. Results for different values of $U_{fc}/D$ lie on the same line, exhibiting a universal behavior.

4.2 Superconducting Transition Temperature

Next, we discuss a possible type of the superconducting gap near the region where the rapid valence change occurs owing to the f-c Coulomb interaction in the extended periodic Anderson model. In the conventional PAM, problems of determining the superconducting transition temperature have been studied by several authors within the slave-boson and $1/N$-expansion method.$^{29,30}$ Following their method, we calculate the superconducting transition temperature in the weak coupling theory in the present framework. The transition temperature is obtained from the integral equation of the particle-particle scattering amplitude, $\Gamma$, for two quasiparticles with opposite momentum near the Fermi surface. (See Fig. 4.) The quasiparticle operators can be represented by a
Fig. 4. Feynman diagram representing the integral equations for the scattering amplitude of two quasiparticles \( \Gamma \) from \( (k, -k) \) to \( (k', -k') \).

Fig. 5. Feynman diagram for the irreducible vertex part \( \Gamma^{(0)} \) to leading order in \( 1/N \). The explicit form of the fluctuation propagators \( D \) is given by eq. (3.29) and its interaction vertices \( g \) with quasiparticle is shown in Fig. 1.

The unitary transformation in terms of f- and conduction electron operators as

\[
\begin{pmatrix}
\gamma_{k}^{(+)} \\
\gamma_{k}^{(-)}
\end{pmatrix} = \begin{pmatrix}
u_{k} & u_{k} \\
-v_{k} & u_{k}
\end{pmatrix} \begin{pmatrix} i_{k} \\
c_{k}
\end{pmatrix},
\]

where \( \gamma^{(\pm)} \) corresponds to the eigen values \( E^{\pm} \), eq. (4.2), respectively. The scattering amplitude \( \Gamma \) is obtained from two-quasiparticle correlation function,

\[
\langle T_{\tau_{1}} \gamma_{k',m'}^{(-)}(\tau_{1}) \gamma_{k',m'}^{(-)}(\tau_{2}) \gamma_{k,m}^{(-)}(\tau_{3}) \gamma_{k,m}^{(-)}(\tau_{4}) \rangle,
\]

by removing the external legs.

To leading order in \( 1/N \), irreducible vertex part \( \Gamma^{(0)} \) includes only a single-boson exchange process as shown in Fig. 5. With the use of the relations, eq. (4.5), eq. (3.29), and interaction vertices shown
in Fig. 1, analytic expression of $\Gamma^{(0)}$ is given as follows:

$$\Gamma^{(0)} = v^4 \left\{ D_{\lambda\lambda} + \frac{U_{fc}}{2} D_{\varphi^c\varphi^c} + \frac{U_{fc}}{2} D_{\varphi^c\varphi^c} + \left( \frac{U_{fc}}{2} \right)^2 D_{\varphi^c\varphi^c} \right\} - 4uv^3 \left\{ V D_{\lambda\rho} + \frac{U_{fc}}{2} D_{\varphi^c\varphi^c} \right\}$$

$$+ u^2 v^2 \left[ 2 \left\{ \frac{U_{fc}}{2} D_{\lambda\varphi^f} + \left( \frac{U_{fc}}{2} \right)^2 D_{\varphi^c\varphi^f} \right\} + 4V^2 D_{\rho\rho} \right]$$

$$- u^3 v^4 \frac{U_{fc}}{2} D_{\varphi^f\rho} + u^4 \left( \frac{U_{fc}}{2} \right)^2 D_{\varphi^f\varphi^f}. \tag{4.6}$$

In the weak coupling limit, in which the external momenta are set on the Fermi surface, i.e. $|\mathbf{k}|, |\mathbf{k}'| \to k_F$, and the static limit, $\omega \to 0$, is taken in the boson propagator, the linearized gap equation represented by Fig. 4 can be solved by a conventional method. Namely, the scattering amplitude is decomposed into the Legendre polynomial as

$$\Gamma(\mathbf{k}, \mathbf{k}') = \sum_{\ell=0}^{\infty} (2\ell + 1) \Gamma_{\ell} P_{\ell}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}'). \tag{4.7}$$

The scattering amplitude $\Gamma_{\ell}$ corresponding to the channel with relative angular momentum $\ell$ is given by

$$\Gamma_{\ell} = \frac{\Gamma^{(0)}_{\ell}}{1 + \rho(\mu) \ln(\frac{T_K}{T})}, \tag{4.8}$$

where $\Gamma^{(0)}_{\ell}$'s are related to $\Gamma^{(0)}(\hat{\mathbf{k}} \cdot \hat{\mathbf{k}}')$'s by the formula

$$\Gamma^{(0)}_{\ell} = \frac{1}{2} \int_{-1}^{1} d(cos \theta) \Gamma^{(0)}(\theta) P_{\ell}(cos \theta). \tag{4.9}$$

Then the transition temperature $T_c$ for $\ell$-wave channel is given by

$$T_c = T_K \exp \left[ \frac{1}{\rho(\mu) \Gamma^{(0)}_{\ell}} \right]. \tag{4.10}$$

Here it is noted that the energy cut-off, corresponding to the Debye frequency, is given by $T_K = \bar{\epsilon}_\ell - \mu$, the bandwidth of quasiparticles.
The transition temperature \( T_c \) so calculated can exist only for the \( d \)-wave (\( \ell = 2 \)) channel as far as the channels, \( \ell = 0, 1 \) and 2, are concerned. In Fig. 6 we show \( T_c \) as a function of \( \epsilon_f \), the f-electron level relative to the Fermi level, for several values of \( U_{fc} \), the repulsion between f- and conduction electrons. Parameters adopted are the same as those in Fig. 2. There exists a sharp peak of \( T_c \) at around \( \epsilon_f \) where \( n_f \) starts to show a rapid decrease. Its tendency becomes more drastic as \( U_{fc} \) increases making the valence change sharper. In the region where the f-electron number is decreased enough, \( T_c \) is strongly suppressed. We have investigated which term in eq. (4.6) plays important role for pairing interaction. It turned out that the major part of \( \Gamma^{(0)} \) is induced by the scattering process \((f,f)\rightarrow(f,c)\) or \((f,c)\rightarrow(f,f)\), in which the valence of f-electrons is changed directly. (See fig. 7.)

Fig. 7. \( \Gamma^{(0)}(q) \) as a function of momentum transfer \( q \) for each scattering channel included in eq. (4.6). For example, “ffff” means the \( v^4 \) term of eq. (4.6). The others are represented similarly.

In the Kondo limit where \( \bar{n}_f \approx 1/2 \), the spin fluctuations are believed to play the most important role for the occurrence of superconductivity. In such a region we have to take into account the higher order term beyond \( 1/N \) to discuss the instability to the superconducting state, since the spin-fluctuation contribution to the effective interaction appears only beyond at the order \( (1/N)^2 \). However, the present approach of the order of \( 1/N \) is still expected to work in the region where the valence fluctuations play an important role.

§5. Conclusions and Discussions

We have developed a microscopic theory to support the idea that the rapid change of f-valence of Ce ion is the origin of anomalous behaviors of CeCu$_2$Ge$_2$ under the pressure \( P \approx 17 \) GPa, such as the coincidence of peaks of \( T_c \) and \( \rho_0 \), and the rapid decrease of the coefficient \( A \) of \( T^2 \)-term of the resistivity. To this end, we analyzed the properties of an extended periodic Anderson model with the f-c Coulomb repulsion \( U_{fc} \) by the slave-boson and \( 1/N \)-expansion method. It turned out that such a model indeed contains the ingredient making rapid change of \( n_f \), f-electron number per
site, possible. It was also shown that the superconducting transition temperature of d-wave pairing is sharply enhanced just before the rapid decrease of \( n_f \) as \( \epsilon_f \) is increased. It turned out that the scattering process of \((f,f) \rightarrow (f,c)\) or \((f,c) \rightarrow (f,f)\), in which the f-electron number is changed directly, plays a major part in Cooper pair formation. Cooper pairing of channels other than the d-wave cannot be possible so long as the pairing interaction discussed in the present paper is concerned.

A few remarks should be made concerning the following points. First, although we did not mention the relation between the coefficient \( A \) of \( T^2 \)-term of the resistivity and the Sommerfeld constant \( \gamma \) within the present approach, we have verified that the Kadowaki-Woods relation is reproduced in the heavy electron limit.\(^{32}\) However, the crossover of the universal ratio \( A/\gamma^2 \) between classes of the heavy electron and of the transition metal was not reproduced. We have cognizance of the necessity of calculations in which the dynamical effect of quasiparticle selfenergy is fully taken into account on the order of \( 1/N \). However, we have to leave such calculations for a future study because we need much more considerable amount of study for performing its program.

Second, there exist several points of view different from ours for explaining the anomalous properties of CeCu\(_2\)Ge\(_2\). One of them is to attribute its origin to the orbital fluctuations in the multiband periodic Anderson model, in which the broad bandwidth under high pressure is expected to change the degeneracy of the f-electron state. Indeed this has been proposed in ref. 4 paying attention to the fact that, at the pressure corresponding to the peak of \( T_c \) and \( \rho_0 \), the two temperatures \( T_{1\text{max}} \) and \( T_{2\text{max}} \) at which the resistivity takes peak merges with each other, indicating the Kondo temperature becomes of the order of the crystal field (CF) splitting. The orbital fluctuation mechanism has also been proposed as a possible mechanism for explaining the phenomena observed in CeCu\(_2\)Ge\(_2\), and discussing that the orbital fluctuation is enhanced where \( T_K \) is comparable to CF splitting.\(^{33}\) However, it is not clear whether all the anomalies observed in CeCu\(_2\)Ge\(_2\) can be explained by those mechanisms.

Third, the origin of superconductivity around the QCP is believed to be due to the enhanced spin fluctuations, while we have neglected here its effect because we are interested in the physics in the region far from the QCP. Therefore, the primary origin of the superconducting transition may change to the spin-fluctuation mechanism as QCP is approached. In order to discuss this crossover in the present model, we have to study the problem up to \((1/N)^2\) which is beyond scope of this paper.

Last, we have discussed the superconducting transition temperature in the weak coupling theory. However, it is suggested that the frequency dependence of the selfenergy is important as can be seen in the discussions concerning the Kadowaki-Woods relation.\(^{32}\) So, the study in the strong coupling theory of superconductivity is desired for more solid conclusion. Nevertheless, the result does not seem to be modified qualitatively, judging from our past experience in the problem of Cooper pair formation in isotropic two-dimensional Fermions with short range repulsion.\(^{34}\)
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Appendix A

Here we discuss how the two kinds of boson fields are introduced in relation to the f-c Coulomb interaction. The f-c Coulomb interaction term in eq. (3.1) can be rewritten as

$$\sum_{\alpha\beta} n_{i\alpha}^f n_{i\beta}^c = \frac{1}{4} \sum_i \left[ \left( \sum_{\alpha} (n_{i\alpha}^f + n_{i\alpha}^c) \right)^2 - \left( \sum_{\alpha} (n_{i\alpha}^f - n_{i\alpha}^c) \right)^2 \right]. \quad (A.1)$$

With the use of the identity

$$\int D(\eta \zeta) \exp \left[ \frac{U_{fc}}{4N} \int_0^\beta d\tau \left\{ \sum_{\alpha} \left( n_{i\alpha}^f - n_{i\alpha}^c \right)^2 - \sum_{\alpha} \left( n_{i\alpha}^f + n_{i\alpha}^c \right)^2 \right\} \right] = \text{Const.} \quad (A.2)$$

and eq. (A.2), the term including $U_{fc}$ in eq. (3.4) can be expressed as follows:

$$\int D(\eta \zeta) \exp \left[ - \int_0^\beta d\tau \frac{U_{fc}}{4N} \sum_{\alpha\beta} n_{i\alpha}^f n_{i\beta}^c \right]$$

$$= \int D(cc^\dagger ff^\dagger \eta \zeta) \exp \left[ - \int_0^\beta d\tau \left\{ \frac{U_{fc}}{2N} \left( \eta_i^2(\tau) + \zeta_i^2(\tau) \right) \right. \right.$$ 

$$+ \frac{U_{fc}}{2N} \left( \eta_i(\tau) - \zeta_i(\tau) \right) \sum_{\alpha} n_{i\alpha}^f + \left( \eta_i(\tau) + \zeta_i(\tau) \right) \sum_{\alpha} n_{i\alpha}^c \right\} \right] \quad (A.3)$$

$$= \int D(cc^\dagger ff^\dagger \phi^f \phi^c) \exp \left[ - \int_0^\beta d\tau \left\{ -\frac{NU_{fc}}{4} (\varphi_i^f(\tau)\varphi_i^c(\tau) \right. \right.$$ 

$$+ \frac{U_{fc}}{2} (\varphi_i^c(\tau) \sum_{\alpha} n_{i\alpha}^f + \varphi_i^f(\tau) \sum_{\alpha} n_{i\alpha}^c) \right\} \right] \quad (A.4)$$

In the last line, bose fields, $\eta$ and $\zeta$, have been transformed to the new ones, $\varphi_i^f = N(\eta_i + \zeta_i)$ and $\varphi_i^c = N(\eta_i - \zeta_i)$. After carrying out the functional integral over the Fermion fields, we obtain eqs. (3.6) and (3.7).
Appendix B

Here we derive formal expressions of the Gaussian fluctuation part of the action, eq. (3.21), and symmetry property of the polarization functions, eq. (3.27). Of the terms in the part of action, eq. (3.21),

$$- \text{Tr} \ln \tilde{A} = - \text{Tr} \ln (1 - \hat{G}_0 \hat{M}) = \sum_n \text{Tr}(\hat{G}_0 \hat{M})^n,$$

those with \( n = 2 \) give contributions of the Gaussian fluctuation. Namely, the relevant term is expressed as

$$\frac{1}{2} \text{Tr}(\hat{G}_0 \hat{M})^2 = \sum_{kq} \text{tr}\{\hat{G}_0(k) \hat{M}_{k+q} \hat{G}_0(k + q) \hat{M}_{k+q}}\}. \tag{B.1}$$

The explicit form of \( \hat{G}_0 \hat{M} \) is given as

$$\hat{G}_0 \hat{M}(k, k + q) = \frac{T}{\sqrt{N_L}} \left[ \frac{U_{fc}}{2} G_{cc}^0(k) \tilde{\varphi}^f(-q) + VG_{0}^{cf}(k) \tilde{\rho}(-q) + VG_{0}^{cf}(k) \tilde{\lambda}(-q) + U_{fc} \tilde{\varphi}^c(-q) \right]. \tag{B.2}$$

Then, the diagonal components of the product of four matrices in eq. (B.1) are written as follows:

$$[\hat{G}_0 \hat{M}(k, k + q) \hat{G}_0 \hat{M}(k + q, k)]_{(11)} = \frac{T^2}{N_L} \left( \frac{U_{fc}}{2} G_{cc}^0(k) \tilde{\varphi}^f(-q) + VG_{0}^{cf}(k) \tilde{\rho}(-q) \right) \left( \frac{U_{fc}}{2} G_{0}^{cf}(k + q) \tilde{\rho}(q) + VG_{0}^{cf}(k + q) \tilde{\varphi}^f(q) \right),$$

and

$$[\hat{G}_0 \hat{M}(k, k + q) \hat{G}_0 \hat{M}(k + q, k)]_{(22)} = \frac{T^2}{N_L} \left( \frac{U_{fc}}{2} G_{cc}^0(k) \tilde{\varphi}^f(-q) + VG_{0}^{cf}(k) \tilde{\rho}(-q) \right) \left( \frac{U_{fc}}{2} G_{0}^{cf}(k + q) \tilde{\rho}(q) + VG_{0}^{cf}(k + q) \tilde{\varphi}^f(q) \right). \tag{B.4}$$

Substituting these expressions into eq. (B.1) and making rather tedious rearrangements, we obtain eq. (3.25) with the matrix \( \hat{S} \) whose components are defined by eqs. (3.26) and (3.27). Polarization
function Π's in eq. (3.27) are defined as follows:

\[
\begin{align*}
\Pi^c(k, \omega) &\equiv \Pi^{(cc,cc)}(k, i\omega), \\
\Pi^f(k, \omega) &\equiv \Pi^{(ff,ff)}(k, i\omega), \\
\Pi^{cf}(k, \omega) &\equiv \Pi^{(cf,cf)}(k, i\omega), \\
\Pi_1(k, \omega) &\equiv \frac{1}{2}\{\Pi^{(cc,cf)}(k, i\omega) + \Pi^{(cf,cc)}(k, i\omega)\}, \\
\Pi_2(k, \omega) &\equiv \frac{1}{2}\{\Pi^{(ff,cf)}(k, i\omega) + \Pi^{(cf,ff)}(k, i\omega)\}, \\
\Pi_3(k, \omega) &\equiv \frac{1}{2}\{\Pi^{(cc,ff)}(k, i\omega) + \Pi^{(ff,cc)}(k, i\omega)\}.
\end{align*}
\]  

(B.5)

where Π(αβ,γδ) is defined as

\[
\Pi^{(αβ,γδ)}(k, i\omega) = \frac{T}{N_L} \sum_{\mathbf{q}, \nu} G_{0}^{αβ}(\mathbf{q}, i\nu)G_{0}^{γδ}(\mathbf{q} + \mathbf{k}, i\nu + i\omega),
\]  

(B.6)

where α, β, γ and δ stand for c or f. It is noted that Π(αβ,γδ) satisfies the following symmetry relations:

\[
\begin{align*}
\Pi^{(cc,cf)}(k, i\omega) &= \Pi^{(cf,cc)}(k, -i\omega), \\
\Pi^{(ff,cf)}(k, i\omega) &= \Pi^{(cf,ff)}(k, -i\omega), \\
\Pi^{(cc,ff)}(k, i\omega) &= \Pi^{(ff,cc)}(k, -i\omega).
\end{align*}
\]  

(B.7)

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