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Heavy Fermions due to Cooperative Effects of Coulomb and Electron-Phonon Interactions in the Two-Orbital Periodic Anderson Model

Keisuke MITSUMOTO * and Yoshiaki ŌNO[1]

Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan
[1]*Department of Physics, Niigata University, Niigata 950-2181, Japan*

Using the dynamical mean-field theory, we investigate the two-orbital periodic Anderson model including both the Coulomb interaction U between f-electrons and the electron-phonon interaction g which couple the local orbital fluctuations of f-electrons with Jahn-Teller phonons. It is found that the heavy fermion state caused by U is largely enhanced due to the effect of g. In the heavy fermion state for large U and g, both the orbital and lattice fluctuations are enhanced, while the charge (valence) and spin fluctuations are suppressed; this is a nonmagnetic origin of the heavy fermions. The effect of the mass enhancement for the case with twofold-degenerate phonon mode is larger than that for one phonon mode. The obtained results show a possible nonmagnetic origin of the heavy fermion state recently observed in $\mathrm{SmOs_4Sb_{12}}$.

KEYWORDS: heavy fermion, periodic Anderson model, orbital degeneracy, Coulomb interaction, electron-phonon coupling, Jahn-Teller phonon, dynamical mean-field theory

1. Introduction

Filled skutterudite compounds $R\mathrm{Os_4Sb_{12}}$ (R: rare earth) have attracted much attention as they have an unique oversized cage made of Os and Sb atoms in which the R ion is included. $\mathrm{SmOs_4Sb_{12}}$ shows a large specific heat coefficient $\gamma = 820$ mJ/(K$^2\cdot$ mol) which is almost independent of applied magnetic fields,[1)] suggesting a heavy fermion state of nonmagnetic origin such as charge, valence and phonon degrees of freedom.[2–9)]

$\mathrm{PrOs_4Sb_{12}}$ shows a large specific heat coefficient $\gamma = 750$ mJ/(K$^2\cdot$ mol) together with a large jump in the specific heat $\Delta C/T_\mathrm{c} \gtrsim 500$ mJ/(K$^2\cdot$ mol) at the superconducting transition temperature $T_\mathrm{c} = 1.85$ K.[10)] In the ultrasonic measurements, remarkable frequency dependence of the elastic constant (ultrasonic dispersion) around 30 K has been observed in $\mathrm{PrOs_4Sb_{12}}$ and has been attributed to large amplitude local vibrations (rattling) of the Pr ion in the cage.[11)] The ultrasonic dispersion occurs in $(C_{11} - C_{12})/2$ corresponding $\mathrm{E_g}$ representation in the cubic symmetry. In addition, $\mathrm{PrOs_4Sb_{12}}$ shows an anomalous softening of the elastic constant below 10 K down to T_c.[11)] The softening is well accounted for by quadrupole susceptibility for a Γ_1 singlet ground state and a $\Gamma_4^{(2)}$ triplet 1st excited state located at 8 K in the crystalline electric field except for an extra softening below 3 K,[12)] where the coupling between the quadrupole fluctuations and a sort of local phonon may play important roles for the extra softening and also for the heavy fermion behavior down to T_c.

As the strong correlation effect due to both the Coulomb interaction and the electron-phonon coupling is crucial for describing the heavy-fermion state in these systems, we need reliable and nonperturbative approaches such as the dynamical mean-field theory (DMFT).[13)] We have studied the single-orbital periodic Anderson-Holstein model by using the DMFT combined with the exact diagonalization (ED) method.[14–16)] What we found are as follows: (1) In the strong electron-phonon coupling regime, $g \gtrsim g_c$, the system shows an anomalous heavy fermion behavior which is accompanied by a large lattice fluctuation and an extreme phonon softening. (2) A simple harmonic potential for ions for $g \lesssim g_c$ changes into an effective double-well potential for $g \gtrsim g_c$. (3) The pairing interaction between the conduction electrons has a maximum at $g \approx g_c$. (4) The heavy fermion state due to the electron-phonon coupling is realized in the wide range of the f-electron number n_f, while that due to the Coulomb interaction is realized in the narrow range near the half-filling $n_f \sim 1$. (5) The effect of the electron-phonon coupling on the heavy fermion state and that of the Coulomb interaction compete with each other.

Recently, we have also investigated the two-orbital periodic Anderson model, where Jahn-Teller (JT) phonons with a twofold-degenerate phonon mode couple with orbital fluctuations of f-electrons which interact with each other via the Coulomb interaction and hybridize with conduction electrons.[17,18)] What we found are as follows: (1) The local orbital and lattice fluctuations are enhanced, while the local charge (valence) and spin fluctuations are suppressed. (2) The sharp soft phonon mode with a large spectral weight is observed for small U, while the broad soft phonon mode with a small spectral weight is observed for large U. (3) The cooperative effect for half-filling with $n_f = 2$ is more pronounced than that for quarter-filling with $n_f = 1$.

In the present paper, we investigate effects of the Coulomb interaction and the electron-phonon coupling on the heavy-electron states in the two-orbital periodic Anderson model coupled with JT phonons in cases with one and twofold-degenerate phonon mode by using the DMFT.

2. Formulation

Our model Hamiltonian is given by,

$$\begin{aligned} H &= \sum_{\mathbf{k}l\sigma} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}l\sigma} c_{\mathbf{k}l\sigma} + \epsilon_f \sum_{il\sigma} f^\dagger_{il\sigma} f_{il\sigma} \\ &+ V \sum_{il\sigma} (f^\dagger_{il\sigma} c_{il\sigma} + h.c.) \end{aligned}$$

*E-mail address: mitsumoto@phys.sc.niigata-u.ac.jp

$$
\begin{aligned}
&+ \quad U\sum_{il}\hat{n}_{fil\uparrow}\hat{n}_{fil\downarrow}+U'\sum_{i\sigma\sigma'}\hat{n}_{fi1\sigma}\hat{n}_{fi2\sigma'} \\
&+ \quad J\sum_{i\sigma\sigma'}f^{\dagger}_{i1\sigma}f^{\dagger}_{i2\sigma'}f_{i1\sigma'}f_{i2\sigma} \\
&+ \quad g_1\sum_{i}(b^{\dagger}_{i1}+b_{i1})\hat{\tau}_{ix}+g_2\sum_{i}(b^{\dagger}_{i2}+b_{i2})\hat{\tau}_{iz} \\
&+ \quad \sum_{i\nu}\omega_{0\nu}b^{\dagger}_{i\nu}b_{i\nu} \qquad (1)
\end{aligned}
$$

with

$$\hat{\tau}_{ix} = \sum_{\sigma}(f^{\dagger}_{i1\sigma}f_{i2\sigma}+f^{\dagger}_{i2\sigma}f_{i1\sigma}) \qquad (2)$$

$$\hat{\tau}_{iz} = \sum_{\sigma}(f^{\dagger}_{i1\sigma}f_{i1\sigma}-f^{\dagger}_{i2\sigma}f_{i2\sigma}), \qquad (3)$$

where $c^{\dagger}_{il\sigma}$ ($f^{\dagger}_{il\sigma}$) is a creation operator for a conduction (c-)electron (f-electron) with orbital $l(=1,2)$ and spin $\sigma(=\uparrow,\downarrow)$ at site i, and $\hat{n}_{fil\sigma} = f^{\dagger}_{il\sigma}f_{il\sigma}$. The orbital index l distinguishes two Kramers doublets like as Γ_8 orbitals in the $\mathrm{O_h}$ symmetry and Γ_{67} orbitals in the $\mathrm{T_h}$ symmetry. $b^{\dagger}_{i\nu}$ is a creation operator for a Jahn-Teller phonon with mode $\nu(=1,2)$ at site i, where the normal coordinate for the Jahn-Teller phonons is given by $\hat{Q}_{i\nu}=1/\sqrt{2\omega_{0\nu}}(b_{i\nu}+b^{\dagger}_{i\nu})$. We assume the twofold-degenerate phonon mode have $\mathrm{E_g}$ mode in the cubic symmetry. $\epsilon_{\mathbf{k}}$, ϵ_f, and V are the dispersion of c-electron, the atomic level of f-electron, and the c-f hybridization, respectively. In the model eq. (1), the Jahn-Teller phonons with the frequency $\omega_{0\nu}$ couple with the local orbital fluctuations of f-electrons, $\hat{\tau}_{ix}$ and $\hat{\tau}_{iz}$ corresponding to $\mathrm{e_g}$ mode, via the electron-phonon coupling g_ν.[19] We note that the local orbital fluctuations of f-electrons with $\mathrm{e_u}$ mode have $\mathrm{e_u}\otimes\mathrm{e_u}$ mode and include $\mathrm{e_g}$ mode. The model eq. (1) also includes the Coulomb interaction between f-electrons: the intra and inter orbital direct Coulomb U and U' and the exchange coupling J. For simplicity, we assume $U=U'$ and $J=0$. In the case with twofold-degenerate phonon mode, we set $\omega_{01}=\omega_{02}(\equiv\omega_0)$ and $g_1=g_2(\equiv g)$ due to the symmetry. In the case with one phonon mode, we assume $g=g_1\neq 0$ with $g_2=0$ or $g=g_2\neq 0$ with $g_1=0$, where the results of both cases are equivalent due to the symmetry.

To solve the model eq. (1), we use the DMFT in which the model is mapped onto an effective single impurity two-orbital Anderson model coupled with Jahn-Teller phonons.[13,20] The local Green's function $G_{fl\sigma}(\mathrm{i}\omega_n)$ and the local self-energy $\Sigma_{l\sigma}(\mathrm{i}\omega_n)$ for the f-electron satisfy the following self-consistency conditions:

$$
\begin{aligned}
G_{fl\sigma}(\mathrm{i}\omega_n) &= \int d\epsilon\frac{\rho(\epsilon)}{\mathrm{i}\omega_n-\epsilon_f-\Sigma_{l\sigma}(\mathrm{i}\omega_n)-\frac{V^2}{\mathrm{i}\omega_n-\epsilon}} \\
&= [\tilde{G}_{fl\sigma}(\mathrm{i}\omega_n)^{-1}-\Sigma_{l\sigma}(\mathrm{i}\omega_n)]^{-1}, \qquad (4)
\end{aligned}
$$

where $\rho(\epsilon)$ is the density of states (DOS) for the c-electron, $\rho(\epsilon)=\sum_{\mathbf{k}}\delta(\epsilon-\epsilon_{\mathbf{k}})$. In the above equation, $\tilde{G}_{fl}(\mathrm{i}\omega_n)$ is the bare Green's function for the effective impurity Anderson model with $U=g=0$ in an effective medium which will be determined self-consistently. The effective impurity Anderson model with finite U and/or g is solved by using the ED method for a finite-size cluster to obtain $G_{fl\sigma}(\mathrm{i}\omega_n)$ and $\Sigma_{l\sigma}(\mathrm{i}\omega_n)$ at $T=0$.[13–18,20–22] In the present study, we use 4 site cluster and the cutoff of phonon number is set to be 9 for each Jahn-Teller mode. We assume a semielliptic DOS with the bandwidth $W=1$, $\rho(\epsilon)=2\sqrt{1-\epsilon^2}/\pi$, and we set $V=0.1$ and $\omega_0=0.01$. We concentrate our attention on the half-filled case, $n_f=2$, with keeping the particle-hole symmetry with $\epsilon_f=-3U/2$ and the normal state in the absence of magnetic and orbital orders, where $\Sigma_{l\sigma}(\mathrm{i}\omega_n)=\Sigma(\mathrm{i}\omega_n)$.

3. Results

In Fig. 1(a), we plot the renormalization factor $Z=(1-\frac{\mathrm{d}\Sigma(\omega)}{\mathrm{d}\omega}|_{\omega=0})^{-1}$ as a function of g for $U=0$ and 0.6. When $g=0$, the effective mass of the quasiparticle $m^*/m=Z^{-1}$ increases with increasing U and then the heavy fermion state with $m^*/m\gg 1$ is realized in the strong correlation regime. When the electron-phonon coupling increases, the effective mass increases for all values of U resulting in the heavy fermion state due to the cooperative effect of the Coulomb interaction and the electron-phonon coupling. This is a striking contrast to the case with the single-orbital periodic Anderson-Holstein model where the both effects are compete with each other as mentioned before.[14–16] The effective mass m^*/m for twofold-degenerate phonon mode is larger than that for one phonon mode. We note that the convergent solutions are obtained only for $Z\gtrsim 0.03$ due to the finite size effect and plotted only for the corresponding parameter region of g in Fig. 1.

The local orbital fluctuation written by

$$\langle\tau_z^2\rangle=\langle\hat{\tau}_{iz}^2\rangle=\langle(\hat{n}_{fi1}-\hat{n}_{fi2})^2\rangle \qquad (5)$$

with $\hat{n}_{fil}=\sum_\sigma\hat{n}_{fil\sigma}$ as a function of g for several values of U, where $\langle\tau_z\rangle=\langle\tau_x\rangle=0$ in the absence of orbital order. We note that, due to the symmetry of $\mathrm{e_u}$ representation, $\langle\tau^2\rangle=\langle\tau_z^2\rangle=\langle\tau_x^2\rangle$ in the case with the twofold-degenerate phonon mode, while $\langle\tau^2\rangle=\langle\tau_x^2\rangle$ or $\langle\tau^2\rangle=\langle\tau_z^2\rangle$ in the case with the one phonon mode. The local orbital fluctuation $\langle\tau^2\rangle$ for one phonon mode is slightly enhanced than that for twofold-degenerate phonon mode in Fig. 1(b). When $g=0$, $\langle\tau^2\rangle$ increases with increasing U as previously obtained in the two-orbital periodic Anderson model.[23]

The local charge fluctuation of f-electrons, *i.e.*, the valence fluctuation $\langle(\hat{n}_{fi}-\langle\hat{n}_{fi}\rangle)^2\rangle$ is plotted as a function of g for several values of U in Fig. 1(c). When $g=0$, the valence fluctuation decreases with increasing U due to the electron correlation effect. When $g\neq 0$, the valence fluctuation is suppressed or almost constant due to the effect of g in contrast to the case with the single-orbital periodic Anderson model where the valence fluctuation is coupled with local phonons and is enhanced due to the effect of g.[14–16] The decrease of valence fluctuation for twofold-degenerate phonon mode is slightly enhanced than that for one phonon mode.

The local moment, $\langle\mathbf{S}^2\rangle=\langle\hat{\mathbf{S}}_i^2\rangle=\langle\hat{S}_{ix}^2+\hat{S}_{iy}^2+\hat{S}_{iz}^2\rangle$, written by

$$\langle\mathbf{S}^2\rangle=\frac{3}{4}\left(\langle\hat{n}_{fi}\rangle+2\sum_{\sigma}\langle\hat{n}_{fi1\sigma}\hat{n}_{fi2\sigma}\rangle-2\sum_{ll'}\langle\hat{n}_{fil\uparrow}\hat{n}_{fil'\downarrow}\rangle\right) \qquad (6)$$

is plotted as a function of g for several values of U in Fig. 1(d). When $g=0$, $\langle\mathbf{S}^2\rangle$ is enhanced due to the effect of U. When g increases, $\langle\mathbf{S}^2\rangle$ is suppressed due to effects of double-occupation probabilities as shown in ref. 18 (not shown here).

The local lattice fluctuation is defined by $\langle Q_\nu^2\rangle=\langle(\hat{Q}_{i\nu}-$

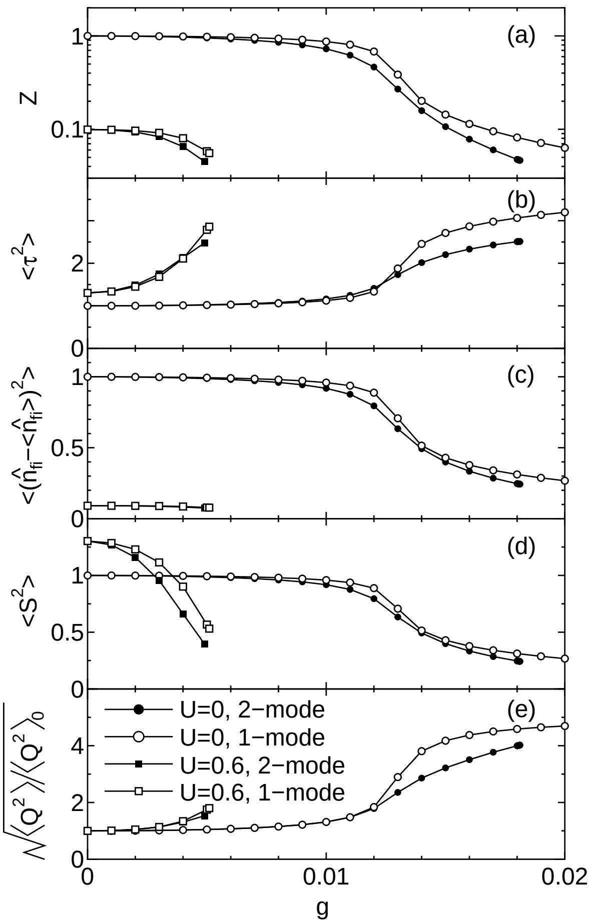

Fig. 1. The renormalization factor Z (a), the local orbital fluctuation $\langle\tau^2\rangle$ (b), the charge fluctuation $\langle(\hat{n}_{fi} - \langle\hat{n}_{fi}\rangle)^2\rangle$ (c), the local moment $\langle\mathbf{S}^2\rangle$ (d), and the root mean squared displacement $\sqrt{\langle Q^2\rangle/\langle Q^2\rangle_0}$ (e) as a function of g for $U = 0$ and 0.6 at half-filling for one (open mark) and twofold-degenerate (solid mark) phonon mode.

$\langle\hat{Q}_{i\nu}\rangle)^2\rangle$ and $\langle Q_1^2\rangle = \langle Q_2^2\rangle (\equiv \langle Q^2\rangle)$ due to the symmetry of E_g modes. In Fig. 1(e), we plot the root mean square displacement, *i.e.*, lattice fluctuation, $\sqrt{\langle Q^2\rangle/\langle Q^2\rangle_0}$, where $\sqrt{\langle Q^2\rangle_0} = 1/\sqrt{2\omega_0}$ is the value for the zero-point oscillation with $g = 0$. When g increases, $\sqrt{\langle Q^2\rangle}$ monotonically increases for all values of U. Remarkably, the increase in $\sqrt{\langle Q^2\rangle}$ observed in the strong coupling regime is largely enhanced due to the Coulomb interaction, where the orbital fluctuation coupled with the local phonons is largely enhanced. This is a striking contrast to the case with the single-orbital periodic Anderson model where $\sqrt{\langle Q^2\rangle}$ is suppressed due to the Coulomb interaction as well as the valence fluctuation as mentioned before.[14–16] When g increases, $\sqrt{\langle Q^2\rangle}$ for twofold-degenerate phonon mode is slightly suppressed than that for one phonon mode.

4. Summary and Discussions

In summary, we have investigated the two-orbital periodic Anderson model coupled with the one and twofold-degenerate Jahn-Teller phonon by using the dynamical mean-field theory. We have found that the heavy fermion state of nonmagnetic origin is realized due to the cooperative effect of the Coulomb interaction U and the electron-phonon coupling g. The specific features of the heavy fermion state for large U and g are as follows: The local orbital and lattice fluctuations are enhanced, while the local charge (valence) and spin fluctuations are suppressed. The effects of mass enhancement for twofold-degenerate phonon mode are larger than that for one phonon mode.

In the two-orbital periodic Anderson model with the coupling g between the local orbital fluctuation and the Jahn-Teller phonon, the orbital fluctuation is enhanced due to the both effects of U and g, and then the heavy-fermion state is realized due to the cooperative effect. This is a striking contrast to the case with the single-orbital periodic Anderson model with the coupling g between the local charge fluctuation and the local phonon, where the spin (charge) fluctuation is enhanced (suppressed) due to the effect of U, while the charge (spin) fluctuation is enhanced (suppressed) due to the effect of g, and then the effects of U and g on the heavy-fermion state compete with each other.[14–16] The heavy fermion state of nonmagnetic origin due to the cooperative effect of U and g seems to be consistent with the magnetically robust heavy fermion state observed in the filled skutterudite $SmOs_4Sb_{12}$.

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