Heavy Fermions in the Periodic Anderson Model with Singlet-Triplet Crystal-Field Levels

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Abstract

We investigate the two-orbital periodic Anderson model with f^2-configuration by using the dynamical mean-field theory in which the effective impurity Anderson model is solved using the exact diagonalization method. The renormalization factor Z and the local moment $\langle \mathbf{S}^2 \rangle$ are obtained as functions of the Coulomb interaction U, the exchange (Hund's rule) coupling J and the crystal-field splitting Δ. When the ionic ground state is triplet for $\Delta < \Delta_c = 3J$, the heavy fermion state with the mass enhancement $m^*/m = Z^{-1} \gtrsim 100$ together with $\langle \mathbf{S}^2 \rangle \sim 2$ is realized in the strong correlation regime for $U \gtrsim W$, where W is the conduction band-width. When the ionic ground state is singlet for $\Delta \gg \Delta_c$, $m^*/m \sim 1$ and $\langle \mathbf{S}^2 \rangle \sim 0$ for any value of U. When the singlet ground state and the triplet excited state form a quasi-quartet for $\Delta \gtrsim \Delta_c$, the system shows a moderate mass enhancement $m^*/m \sim 10 - 100$.

Key words: filled skutterudite, heavy fermion, periodic Anderson model, crystal-field splitting
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Pr-based filled skutterudite compounds have attracted much interest because of the heavy-fermion behavior on the basis of the $4f^2$ configuration of Pr^{3+} ions. $PrFe_4P_{12}$ shows the heavy fermi liquid state with enhanced cyclotron effective mass $m_c^* = 80m_0$ [1]. $PrOs_4Sb_{12}$ exhibits the heavy fermion superconductivity with a large specific heat coefficient $\gamma = 750$mJ/K^{2}mol together with a large jump in specific heat $\Delta C/T_c \sim 500$mJ/K^{2}mol at $T_c = 1.85$K [2]. The crystal-field levels of the Pr^{3+} ion in $PrOs_4Sb_{12}$ are believed to be the ground state Γ_1 singlet and the first excited state $\Gamma_4^{(2)}$ triplet with the excitation energy ~10K [3,4].

To elucidate the effect of the crystal-field levels on the heavy fermion behavior in the f^2-configuration, we study the two-orbital periodic Anderson model at half-filling where the average f-electron number per site is 2. The Hamiltonian of this model is given by

$$H = \sum_{m=1}^{2} \left\{ \sum_{k\sigma} \epsilon_k c_{km\sigma}^\dagger c_{km\sigma} + V \sum_{i\sigma} (f_{im\sigma}^\dagger c_{im\sigma} + h.c.) \right\} + \sum_{m=1}^{2} \sum_{i\sigma} \epsilon_{fm} f_{im\sigma}^\dagger f_{im\sigma} + U \sum_{m=1}^{2} \sum_{i} n_{im\uparrow}^f n_{im\downarrow}^f + \left(U' - \frac{J}{2} \right) \sum_i n_{i1}^f n_{i2}^f - 2J \sum_i \mathbf{S}_{i1}^f \cdot \mathbf{S}_{i2}^f, \quad (1)$$

where the crystal-field splitting is defined by $\Delta = \epsilon_{f2} - \epsilon_{f1}$. We assume a semielliptic DOS for the bare conduction band: $N(\epsilon) = \frac{2}{\pi}\sqrt{1 - (\epsilon/W)^2}$ with the half-bandwidth $W = 1$. For simplicity, we set $U = U' + 2J$ and vary the parameters: U', J and Δ. To solve the model eq.(1), we use the dynamical mean-field theory in which the effective two-orbital impurity Anderson model is solved using the exact diagonalization for a finite size cluster [5]. The present calculations are per-

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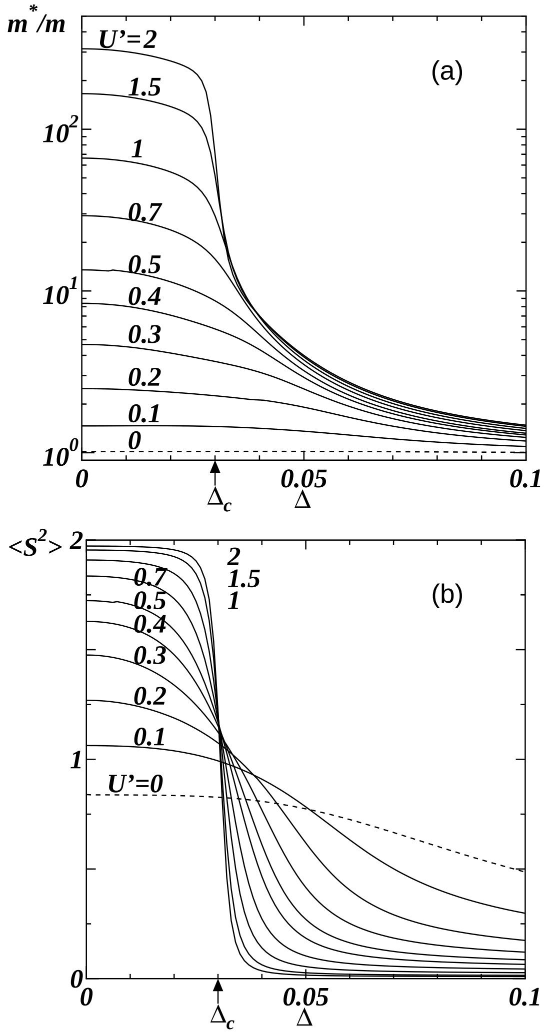

Fig. 1. The mass enhancement factor m^*/m (a) and the local moment $\langle \mathbf{S}^2 \rangle$ as functions of Δ for several values of U' at $J = 0.01$ and $V = 0.1$. When $\Delta < \Delta_c = 3J$, the ionic ground state is triplet, while, when $\Delta > \Delta_c$ it is singlet.

formed at $T = 0$ for 4-site cluster with a fictitious inverse temperature $\tilde{\beta} = 400$ which determines the energy resolution [6].

In the case with isolated ions for the c-f hybridization $V = 0$, the ionic ground state is singlet with $\langle \mathbf{S}^2 \rangle = 0$ when Δ is larger than a critical value $\Delta_c = 3J$, while it is triplet with $\langle \mathbf{S}^2 \rangle = 2$ when $\Delta < \Delta_c$. The quasi-quartet state, where the energy levels for the singlet and the triplet states are almost degenerate, is realized for $\Delta \approx \Delta_c$.

In Figs. 1(a) and (b), the mass enhancement factor $m^*/m = Z^{-1}$ and the local moment $\langle \mathbf{S}^2 \rangle = \langle (\mathbf{S}^f_{i1} + \mathbf{S}^f_{i2})^2 \rangle$ are plotted as functions of Δ for several values of U' at $J = 0.01$ and $V = 0.1$, where the renormalization factor is given by $Z = (1 - \frac{\mathrm{d}\Sigma(\omega)}{\mathrm{d}\omega}|_{\omega=0})^{-1}$. When the ionic ground state is triplet for $\Delta < \Delta_c$, m^*/m increases with increasing U' and finally becomes more than 100 in the strong correlation regime $U' \gtrsim W$. At the same time, $\langle \mathbf{S}^2 \rangle$ increases with increasing U' and finally becomes ~ 2 for $U' \gtrsim W$. On the other hand, when the ionic ground state is singlet for $\Delta > \Delta_c$, with increasing U', m^*/m tends to be a saturated value while $\langle \mathbf{S}^2 \rangle$ decreases down to 0. No distinct transition has been observed in the case with the present model in contrast to the case with the two-orbital Hubbard model in which the discontinuous Mott metal-insulator transition was observed in the strong correlation regime for any finite J [7].

When the ionic ground state is quasi-quartet for $\Delta \approx \Delta_c$, a rapid crossover between the above two regimes is observed for large U as shown in Figs. 1(a) and (b). Especially, in the case of singlet ground state and triplet excited state with a small excitation energy $\Delta - \Delta_c$, the system shows a moderate enhancement of the effective mass $m^*/m \sim 10 - 100$. This is due to the effect of quantum fluctuation between the singlet ground state and the triplet excited state caused by the c-f hybridization. Such effect is considered to play an important role for the heavy-fermion behavior observed in the Pr-based filled skutterudite compounds such as $PrFe_4P_{12}$ and $PrOs_4Sb_{12}$. To discuss the heavy-fermion behavior in such compounds, it is necessary to take account of the spin orbit coupling and the realistic CEF splitting, which are not considered in the present study and will be studied in the future.

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