Staggered Order with Itinerant and Localized Singlets in Generalized Kondo Lattice

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Abstract. The generalized Kondo lattice where even number of localized electrons are coupled with two conduction bands is investigated using the dynamical mean-field theory combined with continuous-time quantum Monte Carlo method. The crystalline electric field (CEF) singlet-triplet states are taken for localized states. With one conduction electron per site, a staggered order with Kondo and CEF singlets arises. Nature of the electronic order is studied in terms of the local susceptibility. The effective CEF splitting in one sublattice remains almost the same as the original CEF splitting, while the other sublattice looks like Kondo singlets. Hence, this electronic order is interpreted as alternating sites of itinerant and localized singlet states.

1. Introduction
Distinction between itinerant and localized characters of electrons is one of the most fundamental problems in strongly correlated electron systems. The typical systems are Lanthanide and Actinide compounds which have nearly localized $f$ electrons. In the system with one $f$ electron per site as in Ce compounds, the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction determines the behavior of the system. As a result of the competition, the quantum phase transition between magnetic and nonmagnetic ground states arises. The overall behavior of this system is now well understood. In the system with two $f$ electrons per site as in some Pr and U compounds, on the other hand, the problem is qualitatively different from $f^1$ systems because of the absence of the Kramers degeneracy.

Some $f^2$ systems can form the crystalline electric field (CEF) singlet ground state in the localized limit of $f$ electrons. If $f$ electrons strongly interact with conduction electrons, the system forms a collective Kondo singlet. In this case, $f$ electrons acquire the itinerancy because of the Kondo effect. In the competing region between Kondo and CEF effects, or itinerant and localized states of $f$ electrons, an exotic ordering is expected. This picture may be relevant to real systems such as PrFe$_4$P$_{12}$ and URu$_2$Si$_2$ [1, 2]. In our previous work, we have demonstrated the existence of the staggered order with the Kondo and CEF singlets (staggered Kondo-CEF singlet order) using the model where the CEF singlet-triplet states interact with the two-band conduction electrons [3].

In the staggered Kondo-CEF singlet ordered phase, the conduction electrons accumulate more on the Kondo-singlet site, and form the charge density wave. The driving force for this order is the effective inter-site repulsion between Kondo singlets, which is naturally understood from the strong coupling limit with respect to the Kondo exchange interaction [4, 5]. In this regard, the staggered Kondo-CEF singlet order is in contrast with the ordinary multipole order.
by the RKKY interaction which is understood from the weak coupling limit. In this paper, we investigate the properties of this electronic order in terms of itinerant and localized characters of $f$ electrons using local susceptibilities.

2. Model and Methods

We take the generalized Kondo lattice model as follows [6]:

$$
\mathcal{H} = \sum_{k\gamma\sigma} (\varepsilon_{k\gamma} - \mu)c_{k\gamma\sigma}^\dagger c_{k\gamma\sigma} + J \sum_{i} S_{i\gamma} \cdot s_{c\gamma i} + \Delta \sum_{i} S_{1i} \cdot S_{2i},
$$

where $\sigma$ ($\gamma$) denotes the spin (orbital) index. $s_{c\gamma i}$ denotes the conduction spin at site $i$ with orbital $\gamma$. $S_{1i}$ and $S_{2i}$ are the localized spins which represent $f$-electron states. As shown in the second term, the conduction electrons with the orbital index $\gamma$ interact with localized spins with the same index by the Kondo exchange interaction $J$. The third term is the interaction $\Delta$ between localized spins at the same site, which tends to form the local pair singlet. This model is called the “two-band singlet-triplet Kondo lattice model” (2BSTKLM) in the following. Figure 1(a) schematically shows this model. Note that the 2BSTKLM with $\Delta = 0$ corresponds to the two-independent Kondo lattice models. Since the localized states can be regarded as the CEF singlet-triplet states with splitting $\Delta$, the word ‘CEF singlet’ is used to represent the local pair singlet in the rest of this paper.

We employ the dynamical mean-field theory (DMFT) for our analysis [8]. The staggered ordered phase can be investigated by extending the DMFT to the two-sublattice system. As the impurity solver, we use the continuous-time quantum Monte Carlo method (CT-QMC).[9, 10, 11] Since the present CT-QMC is free from numerical difficulties such as the negative sign problem, our calculations are highly accurate in the framework of the DMFT. In this paper, we fix the number $n_c$ of conduction electrons per site as $n_c = 1$, which corresponds to the quarter filling with two bands. The conduction bands are taken as $\varepsilon_{k1} = \varepsilon_{k2} = \varepsilon_k$ for simplicity, although the realistic bands are not symmetric. Each conduction electron forms the hypercubic lattice whose density of states is given by

$$
\rho(\varepsilon) = \frac{1}{D} \sqrt{\frac{2}{\pi}} \exp \left( -\frac{2\varepsilon^2}{D^2} \right),
$$

where we take $D = 1$ as a unit of energy. We choose $J = 0.8$ and $T = 0.01$ in this paper.

In the 2BSTKLM at quarter filling, the staggered Kondo-CEF singlet order arises at low temperatures as illustrated in Fig. 1 (b) [3]. The system becomes insulating under the present condition. Since the $\Delta = 0$ case coincides with the two-independent Kondo lattice models, this electronic order connects to the charge density wave found in the Kondo lattice model at
quarter filling [7]. As Δ increases, the ordered phase breaks down by a first-order transition to the homogeneous phase. At T = 0.01, the transition occurs at Δ ≈ 0.42. We note that the present order survives as long as the number of conduction electrons is close to the quarter filling. For J = 0.8 and Δ = 0.2, this range is 0.8 ≲ n_c ≲ 1.3.

3. Magnetic Susceptibilities and Effective CEF Splitting

Let us investigate basic characteristics of the staggered Kondo-CEF singlet ordered phase using local magnetic susceptibilities defined by

\[ \chi_{M}^{\gamma\gamma'} = \int_{0}^{\beta} \left[ \langle T_{\tau}S_{\gamma}^{z}(\tau)S_{\gamma'}^{z} \rangle - \langle S_{\gamma}^{z} \rangle \langle S_{\gamma'}^{z} \rangle \right] d\tau. \tag{3} \]

Under the present condition, the susceptibility has two independent components \( \chi_{M}^{11} = \chi_{M}^{22} \) and \( \chi_{M}^{12} = \chi_{M}^{21} \). In the limit of large Δ, we obtain \( \chi_{M}^{11} = -\chi_{M}^{12} = 1/(2\Delta) \). In the opposite limit of Δ = 0 with Kondo temperature \( T_K \), we have \( \chi_{M}^{11} = 1/(2T_K) \), while \( \chi_{M}^{12} = 0 \) because each spin independently makes the singlet with conduction electrons.

Figure 2 shows the local magnetic susceptibilities computed at T = 0.01 as a function of Δ. The order parameter, which corresponds to the difference of the number of conduction electrons between Kondo- and CEF-singlet sites, is also shown as \( n_{\text{Kondo}} - n_{\text{CEF}} \). In one sublattice, identified as the Kondo-singlet site, \( |\chi_{M}^{12}| \) is always smaller than \( \chi_{M}^{11} \). Furthermore, the value of \( \chi_{M}^{11} \) is almost independent of Δ. In the other sublattice, identified as the CEF-singlet site, the computed results for \( \chi_{M}^{11} \) and \( \chi_{M}^{12} \) are well understood in terms of the effective Hamiltonian \( H_{\text{eff}} = \Delta_{\text{eff}}S_{1} \cdot S_{2} \) on the CEF site. Here \( \Delta_{\text{eff}} \) is the effective CEF splitting. With use of the expression

\[ \chi_{M}^{12}(T = 0) = -\frac{1}{2\Delta_{\text{eff}}}, \tag{4} \]

the effective CEF splitting can be estimated from the local magnetic susceptibility at low temperature. Figure 3 shows \( \Delta_{\text{eff}} \) estimated from \( \chi_{M}^{12} \) at T = 0.01 using (4). Here the straight line represents \( \Delta_{\text{eff}} = \Delta \). For Δ > 0.05, we observe good correspondence between \( \Delta_{\text{eff}} \) and the original CEF splitting Δ. Hence on the CEF-singlet site, the interaction should be weak between the localized states and conduction electrons.
On the other hand, the f electrons at the Kondo-singlet site acquire the itinerancy because of the Kondo effect. Hence, the staggered Kondo-CEF singlet order can be regarded as the staggered order with itinerant and localized singlet states. As is clear from the formation of the CEF-singlet site, this order is a characteristic for the f$^2$ systems. In the strong coupling limit, the conduction electrons accumulate solely on the Kondo sublattice. With finite coupling, the Kondo cloud extends to the CEF sublattice, but charge density wave (CDW) of conduction electrons still remains. Namely, the conduction electrons at the CEF-singlet site is a part of spatially extended Kondo cloud at the Kondo-singlet site, and do not significantly affect the bare CEF character.

This staggered order is responsible for the insulating behavior of the quarter-filled bands. This staggered order is responsible for the insulating behavior of the quarter-filled bands. Figure 1(b) illustrates the electron configurations for the Kondo site and the CEF site in the staggered order. In the doubled unit cell, the total number of itinerant electrons is four, and the two renormalized bands are completely filled. Note that the CEF site does not participate in the conduction. In the absence of the staggered order, the number of electrons is odd per unit cell, and the system becomes metallic.

Finally we consider the deviation from the bare CEF splitting in the region with $\Delta < 0.05$ shown in Fig. 3. Since the case with $\Delta = 0$ corresponds to the two-independent Kondo lattices, $\chi_M^{12}$ should vanish in this limit. Thus $\Delta_{\text{eff}}$ should diverge as $\Delta \to 0$. On the other hand, the difference of $\chi_M^{11}$ between Kondo- and CEF-singlet sites at $\Delta = 0$ comes from the difference of charge density of conduction electrons. The Kondo-singlet site has more conduction electrons, and has thus smaller $\chi_M^{11}$.

4. Summary

The basic characteristic of the staggered Kondo-CEF singlet order at low temperature has been investigated in terms of the local magnetic susceptibilities. Our calculation using the DMFT+CT-QMC successfully characterizes this novel electronic order. The calculated effective CEF splitting at the CEF-singlet site shows good agreement with the original CEF splitting. Hence, the CEF-singlet site is almost decoupled from conduction electrons. This fact allows us to interpret the staggered Kondo-CEF singlet order as the alternating sites of itinerant and localized singlet states.

One of the authors (S. H.) is supported by the global COE program of The Ministry of Education, Culture, Sports, Science, and Technology (MEXT), Japan. This work is partly supported by a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No 20102008) of MEXT, Japan.

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