EFFECT OF ORBITAL DEGENERACY ON TRIPLET SUPERCONDUCTIVITY IN f-ELECTRON SYSTEMS *

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By paying due attention to f-orbital symmetry, we propose a two-orbital Hubbard Hamiltonian with f-electron hopping and Coulomb interactions as an effective model for f electron systems. We analyze the ground state properties of this model by the exact diagonalization technique and discuss a possible mechanism of unconventional superconductivity.

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Novel magnetism and unconventional superconductivity in f electron systems have been central issues in the research field of condensed matter physics. In strongly correlated electron systems such as high-T_c cuprates and organic superconductors, it has been widely recognized that singlet d-wave superconductivity is induced by antiferromagnetic spin fluctuations. In U-based heavy fermion systems, however, URu_2Si_2 may be d-wave superconductor, while UPt_3 is considered to exhibit triplet p-wave pairing. Stabilization of non s-wave pairing is easily understood by the effect of strong correlation, since the cooper pair is composed of a couple of f electrons with heavy effective mass. However, it is still puzzling what is the key issue to determine the symmetry of cooper pair in f electron systems.

For this problem we shed light on orbital degree of freedom inherent in f electrons. In principle seven f orbitals exist, but the number of orbitals should be effectively reduced in solids, due to the combined effects of strong spin-orbit coupling and crystalline electric field (CEF). However, there still remains orbital degree of freedom among the ground-state multiplet. We

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believe that the competition and cooperation among spin and orbital fluctuations should be a key concept to understand the occurrence of either singlet or triplet superconductivity in f electron systems.

In this paper we first discuss Coulomb interaction terms among f electrons by considering faithfully the symmetry of f orbitals for the tetragonal case as a typical example. Second the model is analyzed by the exact diagonalization technique and the ground state phase diagram is obtained. Then, we discuss superconducting properties for some f electron systems.

Let us first consider the model for f electrons in the j-j coupling scheme. Due to the effect of strong spin-orbit coupling, it is enough to consider the j=5/2 sextet and the Hamiltonian is written in the following form:

$$H = -\sum_{\boldsymbol{ia}} T^{\boldsymbol{a}}_{\mu\nu} a^{\dagger}_{\boldsymbol{i}\mu} a_{\boldsymbol{i}+\boldsymbol{a}\nu} + \sum_{\boldsymbol{i},\mu,\nu,\mu',\nu'} I_{\mu\nu,\mu'\nu'} a^{\dagger}_{\boldsymbol{i}\mu} a^{\dagger}_{\boldsymbol{i}\nu} a_{\boldsymbol{i}\nu'} a_{\boldsymbol{i}\mu'} , \tag{1}$$

where $a_{\boldsymbol{i}\mu}$ is an annihilation operator for f electron in the μ-state at site $\boldsymbol{i}$. Note that μ is the z-component of total angular momentum j, running among $-5/2$, $-3/2$, $\cdots$, $5/2$. The first term indicates the f-electron hopping between neighboring sites connected by vector $\boldsymbol{a}$ and the amplitude $T^{\boldsymbol{a}}_{\mu\nu}$ between μ- and ν-states along $\boldsymbol{a}$ direction is calculated by using the Slater's method [1]. The second term denotes Coulomb interactions and the matrix element $I_{\mu\nu,\mu'\nu'}$ is evaluated from the wavefunctions in the sextet. Note that I vanishes unless $\mu+\nu=\mu'+\nu'$. Due to the lack of space, we skip the details for the evaluation of I, but each integral can be expressed by the Slater–Condon parameters [2].

Further we include the CEF effect to reduce the number of orbitals. Under the cubic CEF, the j=5/2 sextet is split into Γ_7 doublet and Γ_8 quartet. In this paper we consider the Γ_8 quartet and after lengthy algebraic calculations, the Hamiltonian is finally reduced to

$$\begin{aligned} H = & -\sum_{\boldsymbol{ia}\sigma\tau\tau'} t^{\boldsymbol{a}}_{\tau\tau'} f^{\dagger}_{\boldsymbol{i}\tau\sigma} f_{\boldsymbol{i}+\boldsymbol{a}\tau'\sigma} - \varepsilon \sum_{\boldsymbol{i}} (\rho_{\boldsymbol{i}a} - \rho_{\boldsymbol{i}b})/2 + U \sum_{\boldsymbol{i}\tau} \rho_{\boldsymbol{i}\tau\uparrow}\rho_{\boldsymbol{i}\tau\downarrow} \\ & + U' \sum_{\boldsymbol{i}} \rho_{\boldsymbol{i}a}\rho_{\boldsymbol{i}b} + J/2 \sum_{\boldsymbol{i},\sigma,\sigma'} \sum_{\tau\neq\tau'} f^{\dagger}_{\boldsymbol{i}\tau\sigma} f^{\dagger}_{\boldsymbol{i}\tau'\sigma'} f_{\boldsymbol{i}\tau\sigma'} f_{\boldsymbol{i}\tau'\sigma} + J' \sum_{\boldsymbol{i}\tau\neq\tau'} f^{\dagger}_{\boldsymbol{i}\tau\uparrow} f^{\dagger}_{\boldsymbol{i}\tau'\downarrow} f_{\boldsymbol{i}\tau\downarrow} f_{\boldsymbol{i}\tau'\uparrow} , \end{aligned} \tag{2}$$

where $f_{\boldsymbol{i}a\uparrow} = \sqrt{5/6}a_{\boldsymbol{i}5/2} + \sqrt{1/6}a_{\boldsymbol{i}-3/2}$, $f_{\boldsymbol{i}a\downarrow} = \sqrt{5/6}a_{\boldsymbol{i}-5/2} + \sqrt{1/6}a_{\boldsymbol{i}3/2}$, $f_{\boldsymbol{i}b\uparrow} = a_{\boldsymbol{i}1/2}$, $f_{\boldsymbol{i}b\downarrow} = a_{\boldsymbol{i}-1/2}$, $\rho_{\boldsymbol{i}\tau\sigma} = f^{\dagger}_{\boldsymbol{i}\tau\sigma} f_{\boldsymbol{i}\tau\sigma}$, and $\rho_{\boldsymbol{i}\tau} = \sum_{\sigma} \rho_{\boldsymbol{i}\sigma\tau}$. Note that σ and τ denote pseudo-spins ($\uparrow$ and $\downarrow$) and pseudo-orbitals (a and b), respectively. In the first term, $t^{\boldsymbol{a}}_{\tau\tau'}$ is given as $t^{\boldsymbol{x}}_{aa} = -\sqrt{3}t^{\boldsymbol{x}}_{ab} = -\sqrt{3}t^{\boldsymbol{x}}_{ba} = 3t^{\boldsymbol{x}}_{bb}$ for x-direction and $t^{\boldsymbol{y}}_{aa} = \sqrt{3}t^{\boldsymbol{y}}_{ab} = \sqrt{3}t^{\boldsymbol{y}}_{ba} = 3t^{\boldsymbol{y}}_{bb}$ for y-direction, respectively [3]. Note that $t^{\boldsymbol{x}}_{aa}$ is taken as an energy unit hereafter. In the second term, the level splitting ε is introduced to include the effect of tetragonal CEF. In the Coulomb interaction terms, U, U', J, and J' denote intra-orbital, inter-orbital, exchange, and pair-hopping interactions, respectively, expressed by the Racah parameters E_k (k=0,1, and 2) for f orbitals [4] as U=E_0+E_1+$2E_2$, U'=E_0+$(2/3)E_2$, J=$5E_2$, and J'=E_1−$(11/3)E_2$. Note the relation of U=U'+J+J', ensuring the rotational invariance in pseudo-orbital space for the interaction part. We believe that the above Hamiltonian reflecting correct symmetry of f orbitals is a minimal model to discuss magnetism and superconductivity in f electron systems with orbital degeneracy.

Let us now discuss the ground state properties of H by using the exact diagonalization technique. For simplicity we consider only the case of n=1 in the two-dimensional square lattice, where n is electron number per site. This situation correponds to Ce-based heavy fermion superconductors with tetragonal crystal structure $CeTIn_5$ (T=Ir, Rh, and Co) [5].

In Figs. 1(a) and (b), we show spin and orbital correlations as a function of ε for U=U'=4, defined as $S(\boldsymbol{q}) = \sum_{\boldsymbol{i},\boldsymbol{j}} e^{i\boldsymbol{q}\cdot(\boldsymbol{i}-\boldsymbol{j})}\langle s_{z\boldsymbol{i}} s_{z\boldsymbol{j}}\rangle$ with $s_{z\boldsymbol{i}} = \sum_\tau(\rho_{\boldsymbol{i}\tau\uparrow} - \rho_{\boldsymbol{i}\tau\downarrow})/2$ and $O_{\tau\tau'}(\boldsymbol{q}) = \sum_{\boldsymbol{i},\boldsymbol{j}} \mathrm{e}^{i\boldsymbol{q}\cdot(\boldsymbol{i}-\boldsymbol{j})}\langle \rho_{\boldsymbol{i}\tau}\rho_{\boldsymbol{j}\tau'}\rangle$. Note here that J and J' are simply neglected, since we consider the case of n=1. For ε<0.5, there is no dominant component in $S(\boldsymbol{q})$, indicating the paramagnetic (PM) phase, while for ε>0.5, $S(\pi,\pi)$ becomes abruptly dominant, strongly suggesting the antiferromagnetic (AFM) phase. This abrupt change is due to the level crossing, but it is instructive to see the orbital correlation, indicating orbital disordered (OD) phase for ε<0.5 and ferro orbital (FO) phase for ε>0.5. By monitoring the change in spin correlation, we can draw the ground-state phase diagram for n=1, as shown in Fig. 1(c). We see OD-PM phase for small U and FO-AFM phase for large U region.

We have obtained the simultaneous onset of spin and orbital ordering, which can be interpreted as the transition between PM and AFM phases controlled by the suppression of orbital fluctuations. Moreover, just around the transition regime between PM and AFM phases, we can expect the appearance of d-wave superconductivity induced by AFM spin fluctuations. In fact, the present model has been analyzed by the random phase approximation, leading to the transition between PM and d-wave superconducting phases with the increase of ε [3]. If we further increase ε, eventually the system becomes the AFM insulating phase and this successive transition agrees with the present results. Here it is stressed that the orbital degree of freedom plays a key role in controling the ground-state property. We believe that this viewpoint is applicable to the appearance of d-wave superconductivity in $CeTIn_5$.

Unfortunately in the present exact diagonalization for n=1, we have not yet obtained indications for triplet superconductivity. We are on the way to clarify the stabilization mechanism of triplet superconductivity. By using

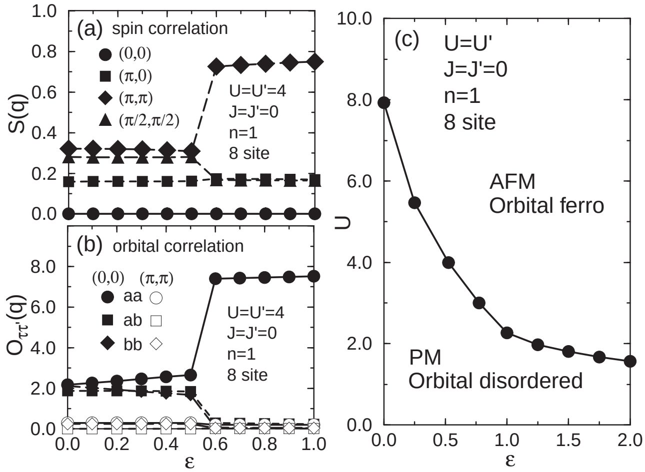

Fig. 1. (a) Spin and (b) orbital correlations as a function of ε. (c) Ground-state phase diagram for n=1 with U=U' and J=J'=0.

the method of *optimized* pair correlation, we have preliminary obtained the dominant triplet pair correlation for the case of n>1 and J>0. In future publications, we will discuss the results in more detail by focusing on the role of Hund's rule coupling.

In summary, we have constructed the model Hamiltonian for f-electron systems and depicted the ground-state phase diagram for n=1, suggesting that unconventional superconductivity appears around the phase boundary between metallic OD-PM and insulating FO-AFM phases.

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