Orbital-Controlled Superconductivity in f-Electron Systems

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We propose a concept of superconductivity controlled by orbital degree of freedom taking CeMIn$_5$ ($M = \text{Co, Rh, and Ir}$) as typical examples. A microscopic multi-orbital model for CeMIn$_5$ is analyzed by fluctuation exchange approximation. Even though the Fermi-surface structure is unchanged, the ground state is found to change significantly among paramagnetic, antiferromagnetic, and $d$-wave superconducting phases, depending on the dominant orbital component in the band near the Fermi energy. We show that our picture naturally explains the different low-temperature properties of CeMIn$_5$ by carefully analyzing the crystalline electric field states.

KEYWORDS: crystalline electric field, heavy-fermion superconductor, fluctuation exchange approximation, thermal expansion

Since the discovery of heavy-fermion superconductivity in CeCu$_2$Si$_2$,$^1$ it has been one of the central issues in the research field of condensed matter physics to unveil unconventional superconductivity in strongly correlated electron systems. In particular, it is important to determine a key parameter for controlling the appearance of superconductivity. Among heavy-fermion superconductors, CeMIn$_5$ ($M = \text{Co, Rh, and Ir}$) have potential for systematic understanding of the mechanism and control parameter of superconductivity, since various ordered phases have been found in the same crystal structure. At ambient pressure, a relatively high superconducting transition temperature $T_c = 2.3$ K has been reported for CeCoIn$_5$, whereas $T_c = 0.4$ K for CeIrIn$_5$. CeRhIn$_5$ is an antiferromagnet with a Néel temperature $T_N = 3.8$ K at ambient pressure, but it becomes superconducting with $T_c \simeq 2$ K at pressure $p \gtrsim 1.6$ GPa.$^4$

The observed power-law temperature dependences of specific heat,$^2,5,6$ thermal conductivity,$^5$ and spin-lattice relaxation rate$^7-10$ suggest the presence of line nodes in the superconducting gap function. In addition, the fourfold magnetic-field angular-dependences of thermal conductivity$^{11}$ and specific heat$^{12}$ clearly indicate $d$-wave superconductivity for CeCoIn$_5$. The existence of antiferromagnetism in CeRhIn$_5$ at ambient pressure is also consistent with $d$-wave superconductivity induced by antiferromagnetic fluctuations.

Concerning the control parameter of superconductivity, one may first consider carrier density, as in the case of high-$T_c$ cuprates. In addition, Fermi-surface topology also plays a crucial role in the occurrence of superconductivity and the determination of the symmetry of the gap function. However, the carrier densities are essentially the same among CeMIn$_5$, because $f$-electron number per Ce ion is not changed by $M$. Furthermore, Fermi surfaces observed by de Haas-van Alphen experiments and band-structure calculations are similar among these compounds.$^{13-18}$ Thus, neither the carrier density nor Fermi-surface topology is the main control parameter of superconductivity and antiferromagnetism in CeMIn$_5$.

Here, we point out another important ingredient, the crystalline electric field (CEF) effect, in CeMIn$_5$. Immediately after the discovery of CeMIn$_5$, Takimoto et al. have stressed the importance of the CEF effect in controlling superconductivity,$^{19}$ but only level splitting was taken into account. Note that CEF potential affects not only level splitting, but also the CEF ground-state wavefunction. In fact, recent inelastic neutron scattering experiments have revealed that level splittings are almost the same among CeMIn$_5$, whereas CEF wave-functions are quite different.$^{20}$ The importance of orbital states has been discussed for Na$_2$CoO$_2\cdot\gamma$H$_2$O,$^{21}$ but the effects of the change in orbital states on superconductivity have not been studied systematically so far. It is conceptually important to clarify superconductivity controlled by orbital states.

In this Letter, we apply fluctuation exchange (FLEX) approximation to an $f$-electron model constructed on a square lattice for CeMIn$_5$. Such perturbative theories have been applied to one-$f$-orbital models for CeMIn$_5$. In this study, we focus on the effect of a CEF using a model including all the states with the total angular momentum $j = 5/2$, which are split into one $\Gamma_6$ and two $\Gamma_7$ doublets under a tetragonal CEF. Among them, the wave functions of $\Gamma_7$ states sensitively depend on CEF potential. We change the wave functions fixing level splittings, and determine both $T_C$ and $T_N$ within FLEX approximation. Even though the Fermi-surface structure does not change, the ground state changes depending on the $\Gamma_7$ wave-functions among the paramagnetic, antiferromagnetic, and $d$-wave superconducting states. We show that the obtained phase diagram is relevant to CeMIn$_5$.

Using the $f$-electron basis under a cubic CEF for convenience, we consider a three-orbital model based on a $j-j$ coupling scheme on a square lattice given by

$$
H = \sum_{\mathbf{r},\mu,\tau,\tau',\sigma} t^\mu_{\tau\tau'} c^\dagger_{\mathbf{r}+\tau\sigma} c_{\mathbf{r}+\tau'\sigma} + U \sum_{\mathbf{r}\tau} n_{\mathbf{r}\uparrow} n_{\mathbf{r}\downarrow} \\
+ (U'/2) \sum_{\mathbf{r},\tau \neq \tau'} n_{\mathbf{r}\uparrow} n_{\mathbf{r}\downarrow} + H_{\text{CEF}},
$$

$1$ where $c_{\mathbf{r}\tau\sigma}$ is the annihilation operator of the $f$ elec-
tron at site $\mathbf{r}$ with the pseudospin $\sigma$ and the orbital $\tau$, $n_{\uparrow\tau\sigma} = c_{\uparrow\tau\sigma}^\dagger c_{\uparrow\tau\sigma}$, and $n_{\downarrow\tau\sigma} = \sum_{\sigma} g_{\downarrow\tau\sigma}$. Note that $\sigma = (\uparrow$ and $\downarrow)$ distinguishes two states in each Kramers doublet. On the other hand, $\tau$ is introduced to distinguish three kinds of Kramers doublets under a cubic CEF. Here, $\alpha$ and $\beta$ denote two $\Gamma_8$, while $\gamma$ indicates $\Gamma_7$. We use $t_{\tau'\tau}$ for the hopping amplitude between the $\tau'$ state at site $\mathbf{r} + \mathbf{r}'$ and the $\tau$ state at site $\mathbf{r}$, where $\mathbf{r}$ is a vector connecting nearest-neighbor sites. We consider the hopping through $(ff\sigma)$ bonding, given by $t_{\alpha\alpha} = t_{\alpha\alpha}' = -\sqrt{3}t_{\beta\beta} = -\sqrt{3}t_{\beta\beta}' = \sqrt{3}t_{\gamma\gamma} = \sqrt{3}t_{\gamma\gamma}' = t$ and zero for the other cases, where $t = 0(fff\sigma)/28$ and $a$ is the lattice constant. The coupling constants $U$ and $U'$ denote intra- and inter-orbital Coulomb interactions, respectively. Note that we ignore the Hund’s rule coupling, pair-hopping interaction, and the other interactions for simplicity. Then, owing to the symmetry requirement, we should set $U = U'$. We also note that $t$, $U$, and $U'$ should be regarded as renormalized ones for heavy electrons.

The tetragonal CEF term $H_{\text{CEF}}$ is given by

$$H_{\text{CEF}} = \sum_\tau \left( B^0_2 O^0_{2\tau} + B^0_4 O^4_{4\tau} + B^1_4 O^1_{4\tau} \right),$$

where $B^m_\tau$ is the CEF parameter and $O^m_{\tau\sigma}$ is the Stevens operator equivalent at site $\mathbf{r}$. We can rewrite eq. (2) in the form of a cubic CEF eigenstates as

$$H_{\text{CEF}} = \sum_{\sigma,\tau,\tau'} B^0_{\tau\tau'} \gamma_{\tau\tau'}^\dagger \gamma_{\tau\tau'} \xi_{\sigma\sigma} \chi_{\sigma\sigma} \chi_{\sigma\sigma},$$

where $B^0_{\tau\tau'} = \frac{1}{2} \delta_{\tau\tau'} \left( \begin{array}{cc} 2U^0_{\tau\tau'} & 0 \\ 0 & 2U^0_{\tau\tau'} \end{array} \right)$ and $U^0_{\tau\tau'} = U^0_{\tau'\tau}$. The effective CEF energy levels, while $\theta$ characterizes eigenstates under a CEF potential. Then, the CEF energy levels are $-\Delta^0_{\tau\tau'}/3 - \Delta^0_{\tau\tau'}/2$ for the $\Gamma_1(1)$ states, $-\Delta^0_{\tau\tau'}/3 + \Delta^0_{\tau\tau'}/2$ for the $\Gamma_2$ states, and $2\Delta^0_{\tau\tau'}/3$ for the $\Gamma_6$ states, $c_{\tau\tau'}^{\dagger \sigma \sigma} \xi_{\sigma\sigma} \chi_{\sigma\sigma} \chi_{\sigma\sigma}$, where $|0\rangle$ denotes vacuum.

Now we apply FLEX approximation, which has been extended for multiorbital models. Since $\sigma$ is a conserved quantity in the present model, the Green’s function does not depend on $\sigma$ and is represented by a $3 \times 3$ matrix. In a matrix form, the Dyson-Gorkov equation is given by $G(k) = G(0)(k) + G(0)(k)\Sigma(k)G(k)$, where $G(k)$ is the Green’s function and $G(0)(k)$ is the noninteracting Green’s function. Here, we have introduced the abbreviation $k = (k, \mathbf{n})$, where $k$ is the momentum and $\mathbf{n} = (2n + 1)\pi T$ is the fermion Matsubara frequency with an integer $n$ and a temperature $T$. The self-energy is given by

$$\Sigma_{\tau\tau'}(k) = \frac{T}{N} \sum_{q_1 q_2} V_{\tau_1 \tau_2}(q) G_{\tau_1 \tau_2}(k) \Sigma_{\tau_1 \tau_2}(k),$$

where $N$ is the number of lattice sites, $q = \{q, \omega_m\}$, and $\omega_m = 2m\pi T$ is the boson Matsubara frequency. The fluctuation exchange interaction is given by $V(q) = \frac{3}{2} [U^\alpha \chi^\alpha(q) U^\alpha - U^\alpha \chi^\alpha(q) U^\alpha/2 + U^\alpha] + \frac{1}{2} [U^\alpha \chi^\alpha(q) U^\alpha - U^\alpha \chi^\alpha(q) U^\alpha/2 - U^\alpha]$. The matrices $U^\alpha$ and $U^\alpha$ are given by $U^\tau_{\tau', \tau'} = U^\tau_{\tau', \tau'} = -U^\tau_{\tau', \tau'} = U^\tau_{\tau', \tau'} = 2U^\tau$, where $\tau \neq \tau'$, and the other matrix elements are zero. The spin and charge parts of the susceptibility are given by $\chi(q) = \chi^0(q) (1 - U^\alpha \chi^\alpha(q) U^\alpha)/2$ and $\chi^0(q) = \chi^{00}(q) (1 + U^\alpha \chi^\alpha(q) U^\alpha)/2$, respectively, where $\chi^{00}(q) = -\frac{1}{\mathcal{N}} \sum_\tau G_{\tau\tau}(k + q) G_{\tau\tau}(k)$.

In FLEX approximation without vertex corrections, the magnetic susceptibility is given by

$$\chi_\nu(q) = \frac{1}{2} \sum_{\tau_1 \tau_2 \tau_3 \tau_4} \chi^{\tau_1 \tau_2 \tau_3 \tau_4}(q) J^{\nu}_{\tau_2 \tau_1 \tau_3 \tau_4} J^{\nu}_{\tau_3 \tau_1 \tau_4 \tau_2} \mathcal{S}_{\tau_1 \tau_2 \tau_3 \tau_4} \mathcal{S}_{\tau_1 \tau_2 \tau_3 \tau_4},$$

where $\nu = x, y, z$, and $\mathcal{S}$ are the Pauli matrices, and $J^{\nu}_{\tau_2 \tau_1 \tau_3 \tau_4}$ is the matrix element of the operator of the dipole moment. In this paper, we renormalize $J^\nu$ so that the sum of squares of eigenvalues is unity, for convenience. The linearized equation for the anomalous self-energy $\phi^\tau$ is expressed as

$$\phi^\tau_{\tau_1 \tau_2}(k) = \frac{T}{N} \sum_{k' \tau_3} K_{\tau_1 \tau_2 \tau_3}(k, k') \phi^\tau_{\tau_3 \tau_2}(k'),$$

where $\mathcal{E}$ denotes the pseudospin singlet (S) or triplet (T) pairing state and the kernel is given by $K_{\tau_1 \tau_2 \tau_3}(k, k') = \sum_{\tau_4 \tau_5} V_{\tau_1 \tau_2 \tau_3 \tau_4}(k, k' - k') G_{\tau_4 \tau_5}(k') G_{\tau_5 \tau_4}(k')$. The effective pairing interactions are given by $V^\tau (q) = \frac{2}{\mathcal{N}} [U^\alpha \chi^\alpha(q) U^\alpha + U^\alpha/2 - \frac{1}{2} U^\alpha \chi^\alpha(q) U^\alpha - U^\alpha/2 + U^\alpha] = \frac{1}{2} [U^\alpha \chi^\alpha(q) U^\alpha + U^\alpha/2 - \frac{1}{2} U^\alpha \chi^\alpha(q) U^\alpha - U^\alpha/2]$. The transition temperature of superconductivity is given by the temperature where eq. (5) has a nontrivial solution.

In the following, we show results for a $32 \times 32$ lattice for $U = U' = 4t$, $\Delta^0 = 1.5t$, and $\Delta = t$. The results are not so sensitive to precise values of $\Delta^0$ and $\Delta$ as long as they are in the order of $t$. In the calculation, we use 1024 Matsubara frequencies. The number of $f$ electrons per site is fixed to be one, since we are considering $\text{Ce}^{3+}$ ions. The present model with the CEF parameter $\theta$ is invariant for $\theta = \theta + \pi$. In addition, when $U = U'$, the model is also invariant for $\theta = -\theta$. Thus, it is enough to consider $0 \leq \theta \leq \pi/2$.

Figures 1(a)–1(c) show the magnetic susceptibility $\chi_\nu(q) = \chi_\nu(q, \omega_m = 0)$ for $\theta = 0, \pi/4$, and $\pi/2$, respectively. It is observed that $\chi_{\nu}(q)$ strongly depends on $\theta$. In the following, we explain this dependence from the viewpoint of itinerant and localized orbitals.

First, let us discuss the noninteracting susceptibility $\chi^{00}(q)$ [Figs. 1(d)–1(f)]. In Figs. 1(g)–1(l), we show noninteracting band structures and Fermi-surface curves. Even if $\theta$ is changed, the band that crosses the Fermi level $E_F$ has similar dispersion at $E_F$, leading to an almost the same Fermi surface. Note, however, that in general, $\chi^{00}(q)$ depends on the band structure and is not determined solely by the Fermi surface structure (e.g., nesting property), particularly for multiorbital systems. The nearly flat band composed mainly of the $\gamma$ orbital locates near $E_F$ for small $\theta$, suggesting that electrons near $E_F$ have nearly localized character. Thus, $\chi_\nu(q)$ for $\theta = 0$ is almost flat in momentum space. For small $\theta$, the magnitude of $\chi^{00}(q)$ is large, since $\chi^{00} \sim \rho_0$, where $\rho_0$ is the density of states at $E_F$, and it becomes large when the localized character is strong.
With increasing $\theta$, the localized character is weakened. Thus, $\chi_{\nu}^{(0)}(q)$ shows a structure in $q$ space and becomes smaller in magnitude. In particular, we observe a moderate enhancement around $q = Q \equiv (\pi/a, \pi/a)$ for $\theta = \pi/4$ and $\pi/2$.

Now, we consider the effect of on-site Coulomb interactions. We expect that the Fermi surface is not markedly affected by the Coulomb interaction. First, note that the susceptibility $\chi$ becomes high and sharp, when the Coulomb interactions are large for a moderate value of $\theta$. The susceptibility $\chi$ is large for $\theta = \pi/4$ and $\pi/2$, respectively. The parameters are set at $U = 1.5t$ and $\Delta_{T} = t$.

Now we discuss possible relevance of our results to CeMIn$_5$. For this purpose, it is necessary to estimate $\theta$ for each material. Among the CEF parameters, the sign of $B_4^2$ is quite important, since the orbital state is markedly affected by the sign. Although inelastic neutron scattering is a powerful method of determining CEF energy levels, the sign of $B_4^2$ cannot be determined solely by neutron scattering experiment. Thus, we should perform CEF analysis of a quantity sensitive to $\langle O_2^4 \rangle$, where $\langle \cdots \rangle$ denotes the expectation value in terms of $H_{\text{CEF}}$. Since the mode of charge distribution corresponding to finite $\langle O_2^4 \rangle$ couples to the lattice, thermal expansion is a good quantity for determining the sign of $B_4^2$. Thermal expansion has been measured for CeMIn$_5$, but the analysis focused only on the second-order term $\langle O_2^2 \rangle$. It is necessary to include fourth-order terms to analyze thermal expansion, as in the case of cubic systems.

Figures 3(a)–3(c) show the temperature dependence of $\alpha_2^0$, $\alpha_0^4$, and $-\alpha_4^4$ for single ion under CEF potential deduced from neutron scattering experiment on CeRhIn$_5$. Solid curves denote the values without a magnetic field and dashed curves denote those in a magnetic field $H_z = 18T$ along the c-axis.

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have more precise knowledge on elastic constants and/or
CeRhIn is located in a small-

magnetic-field effect on $\alpha_0$ and $\alpha_4$ is strong, but weak
on $\alpha_4$. From the experimental results, the magnetic-field effect on the thermal expansion coefficient $\alpha_4$ along the $c$-axis for CeRhIn is not so significant as that observed in Figs. 3(a) and 3(b). Rather it looks similar to
that in Fig. 3(c). Moreover, the minimum with a negative value in $-\alpha_4$ shifts toward higher temperatures under a magnetic field, as is experimentally observed in $\alpha_0$ of CeRhIn. Thus, we highly expect that $\alpha_0$ in CeMIn is mainly determined by $\alpha_4$. For CeCoIn and CeIrIn, $\alpha_0$ is always positive at the temperatures reported, in sharp contrast to that for CeRhIn. These observations indicate that the signs of $B_2^f$ for CeCoIn and CeIrIn are the same, but are different from that for CeRhIn.

For further quantitative analysis, it is necessary to have more precise knowledge on elastic constants and/or magnetoelastic coupling, but it is out of the scope of this paper. Rather, we phenomenologically assume $B_2^f > 0$ for CeRhIn. Then, $B_2^f$ is negative for CeCoIn and CeIrIn. By using the CEF parameters deduced from the neutron scattering experiment, we obtain $\theta = 0.16\pi$ for CeRhIn, $\theta = -0.30\pi$ for CeCoIn, and $\theta = -0.38\pi$ for CeIrIn. In Fig. 4, we show a schematic phase diagram using these values of $\theta$ and experimental results for $T_N$ and $T_c$. This is consistent with the theoretically determined phase diagram in Fig. 2. In addition, CeRhIn locates in a small-$\theta$ region, indicating that $f$ electrons in CeRhIn have a relatively localized character, as is suggested from de Haas-van Alphen experiment. Thus, we conclude that the characteristics of CeMIn can be captured by our model and that the main control parameter of antiferromagnetism and superconductivity for CeMIn is $\theta$ which describes CEF wave-functions. To confirm this, it is important to experimentally determine CEF parameters for CeMIn under pressure.

In summary, we have studied the $f$-electron model including all the states with $j = 5/2$ by FLEX approximation. We have found three kinds of ground state, i.e., paramagnetic, antiferromagnetic, and $d$-wave superconducting states, by changing the CEF wave-functions characterized by the parameter $\theta$, even if we fix level splitting. Such phase change originating from the difference in character of $f$-electron orbitals explains well the difference in CeMIn. This finding shows that, in general, orbital character can be a control parameter of superconductivity, in addition to Fermi surface topology and carrier density. The orbital-controlled superconductivity would be realized also in other $f$-electron materials with active orbital degrees of freedom, and the present study will provide an important step toward the investigation of such exotic superconductivity.

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