Charge Kondo effect and superconductivity in the Falikov-Kimball model with pair hopping

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We study the Falikov-Kimball model with the pair hopping between the conduction and localized bands to discuss how the charge Kondo effect is realized. By combining dynamical mean-field theory with the continuous time quantum Monte Carlo method, we clarify that the charge Kondo state survives even at zero temperature and this competes with the charge ordered and s-wave superconducting states. The role of the interorbital repulsion for the superconducting state is also addressed.

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I. INTRODUCTION

Electron valence in the transition-metal and rare-earth ions has attracted interest in the strongly correlated electron systems. Typical examples are the valence skipping phenomena for bismuth and thallium ions in some compounds. In the ions, electron configurations prefer the closed-shell structure in the s orbital and avoid the ionic state with a spin. This should lead to interesting low-temperature properties such as colossal negative thermal expansion in La-doped $BiNiO_3$ [1–3] and superconductivity in K-doped $BaBiO_3$ [4,5]. Moreover, in the PbTe system with nonmagnetic Tl impurities, Kondo-like behavior appears in the resistivity, which is known as the charge Kondo effect [6,7]. The valence skipping phenomenon in d-electron systems has also been suggested [8,9], which stimulates further theoretical investigations on the valence skipping and related phenomena [10–16].

In valence skipping ions, the effective degrees of freedom should be represented by the empty and doubly occupied states for the s orbital. There are two distinct models to describe the valence skip ions. (i) in most theoretical studies, an effective attractive interaction is introduced in the orbital of the ions to mimic the stability of closed-shell configurations [10–14]. Low-temperature properties have been discussed such as the valence transition in La-doped $BiNiO_3$ [15,16], charge ordering and superconductivity in K-doped $BaBiO_3$ [11], and the charge Kondo effect in Tl-doped PbTe [13,14]. (ii) Another mechanism has recently been proposed, where the interband correlations are taken into account [17]. It has been suggested that the charge Kondo effect in the single impurity model is well reproduced by the introduction of the pair hopping between the impurity and conduction bands in addition to the repulsive interaction. On the other hand, as for the periodic system, the ground state remains unclear as well as the finite-temperature properties. In particular, it should be instructive to clarify in the periodic system the possibility of the superconductivity against the charge Kondo state as the pair hopping may induce the superconducting (SC) state, which is trivially realized in the system with the attractive interaction [18].

In this paper, we study the correlated electron system with conduction and localized bands. By considering Coulomb interaction and pair hopping between conduction and localized orbitals, we discuss how the valence skipping phenomena affect low-temperature properties in the bulk system. Here, we use dynamical mean-field theory (DMFT) [19–21] combined with the continuous-time quantum Monte Carlo (CTQMC) method [22,23]. Examining electron configurations, charge correlations, and order parameters, we discuss the stability of the charge Kondo state against spontaneously symmetry-breaking states.

The paper is organized as follows. In Sec. II, we introduce the model Hamiltonian and briefly summarize our numerical method. In Sec. III, calculating various physical quantities, we discuss the role of interorbital repulsion and pair hopping in realizing the charge Kondo, charge ordered, and superconducting states. Then, we determine the phase diagram. A summary is given in the final section.

II. MODEL AND METHOD

We study low-energy properties in strongly correlated electron systems with the localized valence skipping ions. To this end, we deal with the extended Falikov-Kimball model [24,25], where conduction electrons interact with localized ones. This is the natural extension of the impurity model discussed in Ref. [17], and its Hamiltonian is given as

$$H = H_0 + H', \tag{1}$$

$$H_0 = \sum_{ij\sigma} \left(t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \epsilon_d \delta_{ij} n^d_{i\sigma}\right), \tag{2}$$

$$H' = U_{cd} \sum_{i\sigma\sigma'} n^c_{i\sigma} n^d_{i\sigma'} - J_{ph} \sum_i (c^\dagger_{i\downarrow} c^\dagger_{i\uparrow} d_{i\uparrow} d_{i\downarrow} + \text{H.c.}), \tag{3}$$

where $c_{i\sigma}$ ($d_{i\sigma}$) is an annihilation operator of a conduction electron (localized electron) with spin $\sigma(=\uparrow,\downarrow)$. $n^c_{i\sigma}(= c^\dagger_{i\sigma} c_{i\sigma})$ and $n^d_{i\sigma}(= d^\dagger_{i\sigma} d_{i\sigma})$ are the number operators of the conduction and localized electrons at the ith site, respectively. $t_{ij}(= -t\delta_{\langle ij\rangle})$ is the hopping integral of the conduction electrons between the nearest-neighbor sites and ϵ_d is the energy level of the d orbitals. $U_{cd}(J_{ph})$ is the repulsive interaction (pair hopping) between the conduction and localized electrons.

When $J_{ph} = 0$, the system is reduced to the conventional Falikov-Kimball model [24]. In the infinite dimensions [25], the model is exactly solved, and ground-state properties have

been discussed in detail [26]. It is known that, in the presence of the particle-hole symmetry, the interorbital Coulomb interaction suppresses the single occupancy at each orbital and the charge ordered (CO) state is realized at zero temperature.

In the paper, we consider both the interorbital Coulomb interaction and pair hopping between the conduction and localized bands on an equal footing. In the atomic limit ($t_{ij} = 0$) under the particle-hole symmetry, these interactions prefer the electronic configuration with one of two orbitals empty and the other doubly occupied, while no singly occupied states are realized in each orbital. Therefore, the valence skip feature should be captured in our model. To provide more insight, we wish to introduce the pseudospin operators for αth band as

$$I^x_{i\alpha} = \tfrac{1}{2}(\alpha^\dagger_{i\uparrow}\alpha^\dagger_{i\downarrow} + \alpha_{i\downarrow}\alpha_{i\uparrow}), \tag{4}$$

$$I^y_{i\alpha} = \frac{1}{2i}(\alpha^\dagger_{i\uparrow}\alpha^\dagger_{i\downarrow} - \alpha_{i\downarrow}\alpha_{i\uparrow}), \tag{5}$$

$$I^z_{i\alpha} = \tfrac{1}{2}\big(n^\alpha_i - 1\big). \tag{6}$$

Then, the interaction part of the original Hamiltonian can be rewritten as the following Kondo lattice model with anisotropic interactions [27]:

$$H' = 2\sum_i \big[2U_{cd} I^z_{ic} I^z_{id} + J_{ph}\big(I^x_{ic} I^x_{id} + I^y_{ic} I^y_{id}\big)\big]. \tag{7}$$

We wish to note that U_{cd} and J_{ph} yield distinct low-temperature properties. When $|J_{ph}| \ll 2U_{cd}$, the diagonal Ising interactions make the pseudospins antiparallel in the z direction. If one considers the lattice model, the antiferro-type ordered state is realized with the staggered pseudospin moments $\langle I^z_{i\alpha}\rangle \sim (-1)^{i+\delta_\alpha}$, where $\delta_\alpha = 0(1)$ for a conduction (localized) band. This implies that the CO state is realized in the original model. The characteristic quantities are alternating electron densities ρ_c and ρ_d, where $\rho_\alpha = \sum_i (-1)^i n^\alpha_i/N$. In the opposite case with $|J_{ph}| \gg 2U_{cd}$, the pseudospins are on the xy plane with the staggered configuration, e.g., $\langle I^x_{i\alpha}\rangle \sim (-1)^{i+\delta_\alpha}$ due to the in-plane anisotropy in Eq. (7). Then the superconducting state is realized with the staggered pair potential $\langle \alpha_{i\uparrow}\alpha_{i\downarrow}\rangle \sim (-1)^{i+\delta_\alpha}$. When the particle-hole transformation $c_{i\sigma} \to (-1)^i \sigma \tilde{c}^\dagger_{i\sigma}$ is applied, $H(t, U_{cd}, J_{ph})$ is transformed to $H(t, U_{cd}, -J_{ph})$, and the superconducting order parameter is uniform in the model. Therefore, the sign of the pair hopping is essentially irrelevant, and the SC state can be regarded as a conventional s-wave SC state. When $2U_{cd} = J_{ph}$, the system is reduced to the isotropic Kondo lattice model. In the strong-coupling case, the Kondo insulating state is realized with the pseudospin singlet ($\langle \mathbf{I}_c \cdot \mathbf{I}_d\rangle = -3/4$). This implies the existence of the charge Kondo state in our model, which is mainly formed by empty and doubly occupied states.

To study the competition between the SC, CO, and charge Kondo states in the original model, Eq. (3), we make use of DMFT [19–21] in the Nambu formalism [28]. In the framework of DMFT, the lattice model is mapped to an effective impurity model, where local electron correlations are taken into account precisely. The Green function for the original lattice system is then obtained via self-consistency equations imposed on the impurity problem. The noninteracting Green's function in the lattice system is represented as the 2-by-2 matrix,

$$\hat{G}_{0\alpha}(\mathbf{k}, i\omega_n) = [i\omega_n \hat{\sigma}_0 + (\mu - \epsilon_{\alpha\mathbf{k}})\hat{\sigma}_z]^{-1}, \tag{8}$$

where $\hat{\sigma}_0$ is the identity matrix, $\hat{\sigma}_z$ is the z component of the Pauli matrix, $\omega_n = (2n+1)\pi T$ with interger n is the Matsubara frequency, T is the temperature, and μ is the chemical potential. $\epsilon_{\alpha\mathbf{k}}$ is the dispersion relation for the $\alpha(= c,d)$th band, namely, $\epsilon_{c\mathbf{k}} = \epsilon_{\mathbf{k}}$ and $\epsilon_{d\mathbf{k}} = \epsilon_d$. Since there is no hybridization between conduction and localized bands, no interband elements appear in the Green's function [29]. The lattice Green's function is then given by the site-diagonal self-energy as

$$\hat{G}_\alpha(i\omega_n) = \int d\mathbf{k}\big[\hat{G}^{-1}_{0\alpha}(\mathbf{k}, i\omega_n) - \hat{\Sigma}_\alpha(i\omega_n)\big]^{-1}, \tag{9}$$

where the Green's functions and self-energy are represented in the Nambu formalism.

In the following, we use the semicircular density of states $\rho(x) = 2\sqrt{1-(x/D)^2}/\pi D$, which corresponds to an infinite-coordinate Bethe lattice. By using Dyson equations, the self-consistency condition is represented by the Green's function of the conduction bands, as

$$\hat{\mathcal{G}}^{-1}(i\omega_n) = i\omega_n\hat{\sigma}_0 + \mu\hat{\sigma}_z - \left(\frac{D}{2}\right)^2 \hat{\sigma}_z \hat{G}_c(i\omega_n)\hat{\sigma}_z, \tag{10}$$

where $\hat{\mathcal{G}}$ is the noninteracting Green's function in the effective impurity model.

There are various numerical methods to solve the effective impurity problem. To discuss quantitatively how the SC and CO states compete with the charge Kondo state, we use here the CTQMC method [22,23]. In our model, the double expansion technique [30], where the partition function is expanded with respect to both the effective bath and the pair hopping, is efficient to perform Monte Carlo simulations without minus sign problems. In the paper, to discuss how the valence skipping phenomenon is realized, we evaluate the probabilities of empty, singly, and doubly occupied states in each orbital as $\langle e_{i\alpha}\rangle$, $\langle s_{i\alpha\sigma}\rangle$, and $\langle d_{i\alpha}\rangle$, where $e_{i\alpha} = (1-n^\alpha_{i\uparrow})(1-n^\alpha_{i\downarrow})$, $s_{i\alpha\sigma} = n^\alpha_{i\sigma}(1-n^\alpha_{i\bar{\sigma}})$, and $d_{i\alpha} = n^\alpha_{i\uparrow}n^\alpha_{i\downarrow}$, respectively. In the following, we take D as the unit of energy and set $\mu = U_{cd}$ and $E_d = 0$ to discuss low temperature properties in the system with particle-hole symmetric conditions $\langle n_c\rangle = \langle n_d\rangle = 1$. The errors of the obtained data shown in the figures are smaller than their symbol size.

III. RESULTS

We discuss low-temperature properties in the system with itinerant and localized bands. Fixing the interorbital Coulomb interaction as $U_{cd} = 0$, we focus on the effect of the pair hopping in the system to discuss the competition between the charge Kondo and SC states. We first calculate the probabilities of empty, singly, and doubly occupied states to examine the electron configuration in the system. The results are shown in Fig. 1(a). When $J_{ph} = 0$, the system is noninteracting, and the metallic state is realized with $\langle e^c\rangle = \langle s^c_\sigma\rangle = \langle d^c\rangle = 0.25$. The introduction of the pair hopping increases the probabilities of empty and double occupied states, while it decreases those of single occupied states. In the strong-coupling limit, these values $\langle e^c\rangle = \langle d^c\rangle \to 0.5$ and $\langle s^c_\sigma\rangle \to 0$. Similar behavior

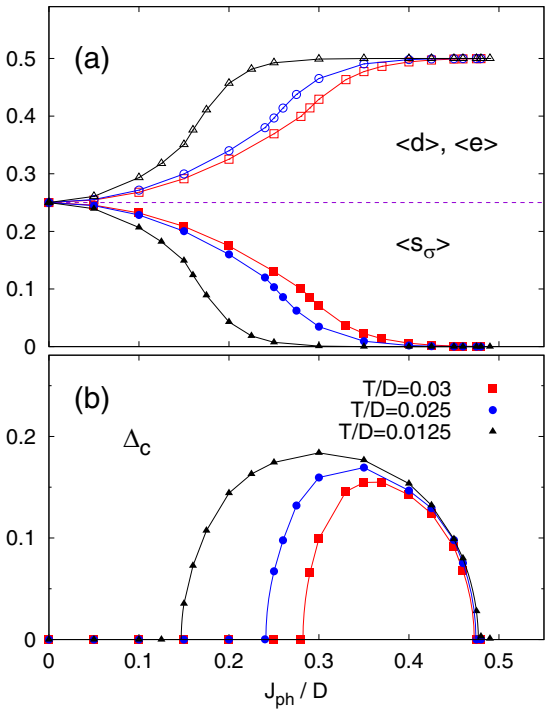

FIG. 1. (a) Electron configurations $\langle d\rangle$, $\langle s_\sigma\rangle$, and $\langle e\rangle$, and (b) the pair potential in the system with $U_{cd} = 0$ at the temperature $T/D = 0.03$, 0.025, and 0.0125, respectively.

appears in the localized bands (not shown), which means that the valence skip behavior is well described by the pair hopping. To discuss how the SC state is realized in the system, we also calculate the pair potential in the conduction band $\Delta_c = \langle c_\uparrow c_\downarrow\rangle$, as shown in Fig. 1(b). In the intermediate-coupling region $(J_{ph})_{c1} < J_{ph} < (J_{ph})_{c2}$, the SC state is realized with a finite pair potential. Namely, the phase transitions are of second order and the critical interactions are deduced as $(J_{ph})_{c1}/D \sim 0.28$ and $(J_{ph})_{c2}/D \sim 0.47$ at $T/D = 0.03$. An important point is that electron configurations are gradually changed in the SC state, as shown in Fig. 1(a). Around $(J_{ph})_{c1}$, the single occupancy still appears because of weak electron correlations. In this case, the BCS-like SC state is realized, and thereby the critical value $(J_{ph})_{c1}$ approaches zero with decreasing temperatures, as shown in Fig. 1(b). Roughly speaking, we find that the emergence of the phase transition appears to be related to the double occupancy of $\langle d\rangle \sim 0.4$, which helps us to discuss later the effect of the interorbital interaction. On the other hand, in the stronger coupling region singly occupied states are little realized, as shown in Fig. 1(a). Therefore, paired electrons formed by pair hopping play a crucial role in the region. When paired electrons are itinerant in the lattice [$J_{ph} < (J_{ph})_{c2}$], the SC state is realized. On the other hand, when $J_{ph} > (J_{ph})_{c2}$, the paired electrons are localized in each site, which is expected to correspond to the charge Kondo state. To clarify whether or not the charge Kondo state is realized at low temperatures, we show in Fig. 2 the temperature dependence of the pseudospin correlation $\langle \mathbf{I}_c \cdot \mathbf{I}_d\rangle$ for the system with $U_{cd} = 0$ and $J_{ph}/D = 0.75$. Its magnitude becomes larger with decreasing temperature at $T \sim |J_{ph}|$. The quantity is almost saturated below $T/D \sim 0.1$, where $\langle e^c\rangle = \langle d^c\rangle \sim 0.5$

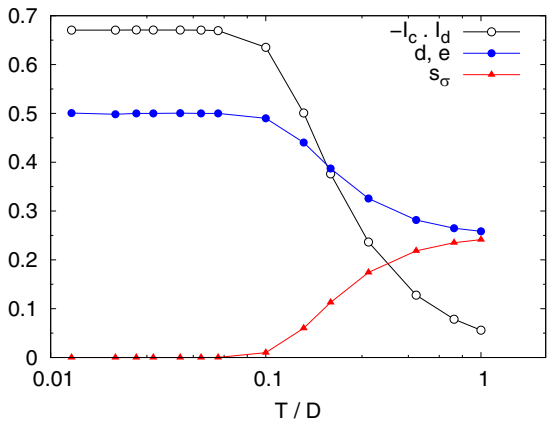

FIG. 2. Pseudospin correlations $\langle \mathbf{I}_c \cdot \mathbf{I}_d\rangle$ and probabilities as a function of the temperature in the system with $U_{cd} = 0$ and $J_{ph}/D = 0.75$.

and $\langle s^c_\sigma\rangle \sim 0$. In addition, we could not find the SC state at lower temperatures, suggesting that the charge Kondo state is realized even at zero temperature. This is consistent with the fact that the critical point $(J_{ph})_{c2}$ between the SC and charge Kondo states is little changed with decreasing temperature, as shown in Fig. 1(b).

We next consider how the interorbital interaction U_{cd} stabilizes the CO state [31–34]. When $J_{ph} = 0$, the system is reduced to a conventional Falikov-Kimball model and its low-temperature properties have been discussed in detail [24,25]. Figure 3 shows the order parameter at fixed temperatures $T/D = 0.03$, 0.05, and 0.1. We find that the CO state is realized in the intermediate-coupling region $U_{cd}/D \sim 1$ and becomes more stable with decreasing temperatures. This is consistent with the fact that the CO state is always a ground state in the system without the pair hopping $J_{ph} = 0$ [31–34].

From these results in two limiting cases, we find two distinct ordered states. Now, another question arises of how the SC and CO states compete with each other. Here, we fix the condition $U_{cd} + J_{ph} = 0.38D$ to clarify how these two

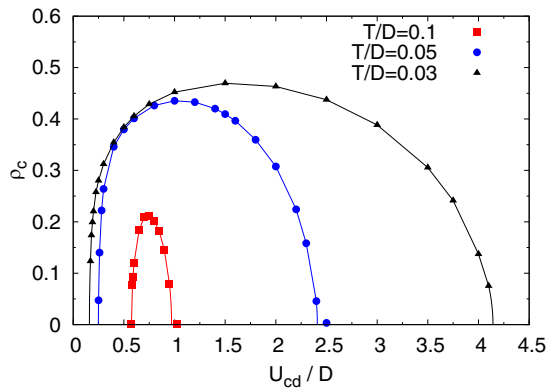

FIG. 3. Solid squares, circles, and triangles represent order parameters for the CO state in the system with $J_{ph} = 0$ at the temperature $T/D = 0.1$, 0.05, and 0.03, respectively.

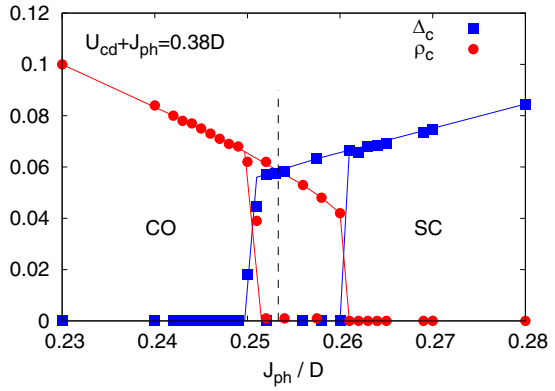

FIG. 4. Circles and squares represent order parameters for the SC and CO states when $U_{cd} + J_{ph} = 0.38D$ and $T/D = 0.03$. Dashed line represents the symmetric point ($J_{ph}/D = 0.2533$).

phases are realized. Figure 4 shows order parameters in the system at $T/D = 0.03$. When $J_{ph}/D = 0.23$, the CO state is realized with the order parameter $\rho_c \sim 0.1$. The increase of the pair hopping monotonically decreases this quantity up to $J_{ph}/D \sim 0.26$. The order parameter suddenly vanishes and the finite pair potential appears instead. This implies the existence of the first-order phase transition between CO and SC states. In fact, the SC state solution exists in the case $J_{ph}/D \gtrsim 0.25$ shown as the solid squares in Fig. 4. Note that at the symmetric point ($2U_{cd} = J_{ph} = 0.2533D$), order parameters take the same value within our numerical accuracy. This originates from the fact that the Hamiltonian Eq. (7) is isotropic and these two states are degenerate at zero temperature. Then, we conclude that the CO and SC states do not coexist in the region $0.25 < J_{ph}/D < 0.26$, and there exists a first-order transition between them.

By performing similar calculations, we obtain the phase diagram with a fixed temperature $T/D = 0.03$, as shown in Fig. 5. When the system is weakly correlated with $U_{cd}, J_{ph} \ll D$, the metallic state is realized. The pair hopping term

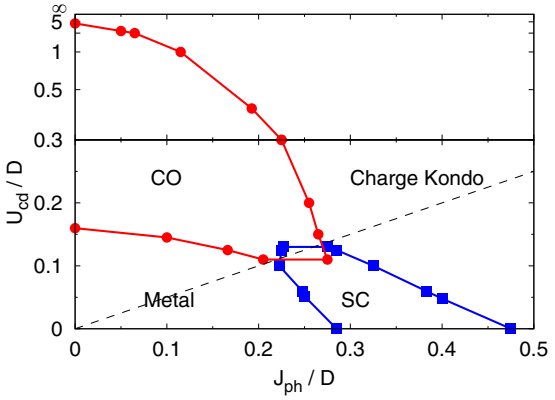

FIG. 5. Phase diagram of the system when $T/D = 0.03$. Solid circles (squares) represent the transition points, where the CO (SC) state disappears. The dashed line represents the symmetric condition ($J_{ph} = 2U_{cd}$). The overlapped area around ($J_{ph}/D, U_{cd}/D$) $\sim$ (0.25,0.125) represents the hysteresis region.

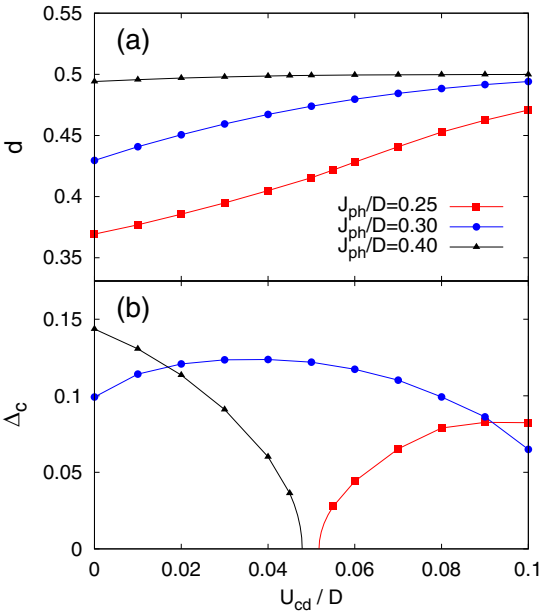

FIG. 6. (a) Double occupancy $\langle d \rangle (= \langle e \rangle = (1 - \langle s_\sigma \rangle))$ and (b) pair potential Δ_c as a function of U_{cd}/D. Solid squares, circles, and triangles represent the results for the systems with $J_{ph}/D = 0.25, 0.3$ and 0.4 at $T/D = 0.03$.

induces the s-wave SC state, while the interorbital interaction induces the CO state. These two solutions overlap around the symmetric line, as discussed above. We also find that the phase boundary for the metallic and SC states becomes lower when the interorbital interaction is switched on. This should be explained by the fact that, in the weak-coupling region, the SC state is induced when empty and doubly occupied states become dominant. In fact, when $J_{ph}/D = 0.25$, the introduction of the interorbital Coulomb interaction increases the double occupancy while it suppresses single occupancy, as shown in Fig. 6(a). We find that the SC state appears around $U_{cd}/D \sim 0.052$ ($\langle d \rangle \sim 0.4$) as shown in Fig. 6(b). This result indicates that the SC state is induced by U_{cd} in the fixed J_{ph}. The pair coupling J_{ph} trivially stabilizes the SC state as the order parameter $\Delta_c = \langle c_\uparrow c_\downarrow \rangle$ is simply obtained by its mean-field decoupling. On the other hand, a static mean-field approximation of the interorbital repulsion term U_{cd} never yields the SC order parameter $\Delta_c = \langle c_\uparrow c_\downarrow \rangle$. Therefore, we can say that dynamical correlations play an important role in stabilizing the SC state in the system. This is similar to the case in the two-orbital Hubbard model [35], where the interorbital repulsive interaction induces the SC state within DMFT. As well as this previous study, since the present calculations are performed by DMFT, critical phenomena always belong to a mean-field universality class and thereby critical exponents are not changed. These discussions are also applied to the CO state in the weak-coupling region, where the pair hopping J_{ph} play a role in forming paired electrons.

In the strong-coupling region with $U_{cd}, J_{ph} \gg D$, the paramagnetic state appears in the phase diagram. Decreasing temperatures, the state is adiabatically connected to the charge Kondo state. In the large U_{cd} case, the characteristic energy for the CO state is D^2/U_{cd}, while that for charge Kondo state

is the pair hopping J_{ph}. Therefore, the ground-state phase boundary between the CO and charge Kondo states should be scaled as $\sim U_{cd}J_{ph}/D^2$ and infinitesimal J_{ph} induces the charge Kondo state in the $U_{cd} \to \infty$ limit. On the other hand, in the $U_{cd} = 0$ case, the charge Kondo state competes with the SC state and its phase boundary is at finite J_{ph} in the ground state, as discussed before. Switching U_{cd} enhances the pseudospin correlations, stabilizing the charge Kondo state. Therefore, the introduction of the interorbital Coulomb interaction makes the strong-coupling SC state unstable, which is clearly found in the case $J_{ph}/D = 0.4$ in Fig. 6(b). Then, the linear behavior in the phase boundary between the SC and charge Kondo states appears in the strong-coupling region. On the other hand, in the weak-coupling region, our DMFT calculations suggest that the metallic phase disappears at $T = 0$. Then, the CO and SC states compete with each other, and its phase boundary should be determined by the symmetric condition $J_{ph} = 2U_{cd}$.

Before closing this paper, we comment on the effect of the single electron hopping (hybridization) between the conduction and localized band, which has been treated in the periodic Anderson model. Since the self-consistency condition Eq. (10) is not changed [36], one can treat this model in the same framework to discuss the possibility of the magnetically ordered state and competition between magnetic and charge Kondo states [37]. We expect that the single hopping activates singly occupied states and the nature of the valence skip ions becomes obscure. However, this hopping gives rise to minus sign problems in solving the effective impurity model by means of the CTQMC method. Therefore, the quantitative analysis should be restricted at relatively higher temperatures. Moreover, it is also interesting to consider additional interactions such as the Hund coupling, which might result in the triplet SC state [38], but it is beyond the scope of our paper. Therefore, these issues are left for future work.

IV. CONCLUSION

We have investigated the extended Falikov-Kimball model with the Coulomb and pair hopping between the conduction and localized bands to discuss how the valence skipping ions induce a spontaneously symmetry-breaking state. By combining DMFT with the CTQMC method, we have determined the finite-temperature phase diagram, where the SC and CO states compete with the charge Kondo state. It is found that, in the weak-coupling region, the Coulomb interaction assists the stability of the SC state, which is a common feature inherent in the multiorbital systems.

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