Two-orbital Systems with Crystal Field Splitting and Interorbital Hopping

Yun Song

Department of Physics, Beijing Normal University, Beijing 100875, China

Liang-Jian Zou

Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, P. O. Box 1129, Hefei 230031, China

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The nonequidistant two-orbital Hubbard model is studied within the dynamic mean-field theory to reveal the influence of two important factors, i.e. crystal field splitting and interorbital hopping, on orbital selective Mott transition (OSMT) and realistic compound Ca$_{2-x}$Sr$_x$RuO$_4$. A distinctive feature of the optical conductivity of the two nonequidistant bands is found in OSMT phase, where the metallic character of the wide band is indicated by a nonzero Drude peak, while the insulating narrow band has its Drude peak drop to zero in the mean time. We also find that the OSMT regime expands profoundly with the increase of interorbital hopping integrals. On the contrary, it is shown that large and negative level splitting of the two orbitals diminishes the OSMT regime completely. Applying the present findings to compound Ca$_{2-x}$Sr$_x$RuO$_4$, we demonstrate that in the doping region from $x = 0.2$ to $2.0$, the negative level splitting is unfavorable to the OSMT phase.

I. INTRODUCTION

The orbital degree of freedom is one of the key points to understand the rich and complicated properties of transition metal oxides, and its roles in Mott-Hubbard metal-insulator transition (MIT) have attracted much interest in recent years. The orbital selective Mott transition (OSMT) in multi-orbital correlated systems, where the Mott-Hubbard metal-insulator transition (MIT) in different orbitals happens successively with the variation of doping, interaction, or field, was proposed to interpret the two consecutive phase transitions and spin state transition in the layered perovskite Ca$_{2-x}$Sr$_x$RuO$_4$. Besides, it was also applied to other orbital compounds, such as V$_2$O$_3$ and VO$_2$. However, this OSMT scenario has not been verified by experiments in the OSMT candidate Ca$_{2-x}$Sr$_x$RuO$_4$ till now. The optical spectra and the angle-resolved photoelectron spectroscopy (ARPES) experiments were performed to examine the electronic states and spectral properties of Ca$_{2-x}$Sr$_x$RuO$_4$, but neither considerable contraction of Fermi surface volume nor the OSMT gap for narrow $d_{xz/yz}$ bands was found near $x = 0.5$. Whereas the most recent result obtained by ARPES experiment on Ca$_{2-x}$Sr$_x$RuO$_4$ at $x = 0.2$ presented a different scenario, where the low-energy quasiparticle excitations in wide $d_{xy}$ orbital disappeared completely.

Great theoretical efforts have been paid to unearth the essential of the OSMT by the dynamical mean-field theory (DMFT) and some other approaches. It has been confirmed that the two successive MITs and the OSMT are robust features in the two-orbital and three-orbital Hubbard models with asymmetric bandwidths. Meanwhile, the essential aspects of the OSMT are not well understood. For example, the effect of Hund’s coupling in OSMT is still not clear. Koga et al. and Liebsch et al. found that the full Hund’s coupling is crucial for the existence of OSMT in two-orbital Hubbard model with asymmetric bandwidth using the DMFT with quantum Monte Carlo solver and exact diagonalization solver, respectively. However, utilizing the slave spin mean-field theory and DMFT, Medici et al. suggested that the OSMT exists over a wider range of $J$ when the ratio of the narrow and wide bandwidths is small enough, even in the full SU(4) spin-orbital symmetric case with $J = 0$. On the other hand, Liebsch et al. recently introduced a three-orbital Hubbard model with positive crystal field splitting for the real compound Ca$_{2-x}$Sr$_x$RuO$_4$, and found a metallic-band insulator transition for the completely filled $d_{xy}$ orbital and a Mott transition in the remaining half-filled $d_{xz/yz}$ bands, precluding the possibility of the OSMT phase in Ca$_{2-x}$Sr$_x$RuO$_4$.

In order to get a clear picture about the unconventional MITs in compound Ca$_{2-x}$Sr$_x$RuO$_4$, all major essentials of the real compound should be taken into consideration when a realistic model is set up. In layered perovskite Ca$_{2-x}$Sr$_x$RuO$_4$, the nearest-neighbor (NN) interorbital hopping integrals are comparable with that of the intraorbital integrals in the order of magnitude. However, in previous studies, the NN interorbital hopping integrals were neglected so as to make the self-energy and Green’s function be diagonal in orbital space. On the other hand, only the hybridization of the two orbitals at the same site was considered recently in reference [11,17]. It is expected that the presence of the interorbital hopping will profoundly affect OSMT. Moreover, both the experiments and the first principle calculations demonstrated that with the increase of doping in Ca$_{2-x}$Sr$_x$RuO$_4$, the structure distortions arising from the rotation and tilting of RuO$_6$ octahedra play an important role in the two successive MITs. As we will analyze later, such distortions lead to the decline of the level center of the narrow band with respect to the wide one, giving rise to a negative crystal field splitting.

The aim of this paper is to elucidate the combining...
effect of two important factors, i.e. interorbital hopping and crystal field splitting, on the optical conductivity and the OSMT in multiorbital correlated model and realistic multiorbital material Ca$_{2-x}$Sr$_x$RuO$_4$. We study the optical conduction of two-orbital Hubbard model within the framework of the extended linearized dynamical mean field theory (ELDMFT)\cite{19}, which has been proved extensively to be an efficient method, especially for the complicated multiorbital models. We demonstrate how the interorbital hopping and the negative crystal field splitting affect the nature of OSMT and the optical conductivity of the systems. We find that the OSMT regime enlarges in the presence of the interorbital hopping, but the negative crystal field splitting strongly restrains the occurrence of OSMT. In addition, the unconventional MITs in compound Ca$_{2-x}$Sr$_x$RuO$_4$ is also discussed. The rest of the paper is organized as follows. In Sec. II, we describe the model Hamiltonian and outline the ELDMFT method; in Sec. III, we present the major theoretical results of the interorbital hopping and the crystal field splitting on the OSMT, and the possible application on compound Ca$_{2-x}$Sr$_x$RuO$_4$. Sec. IV is devoted to the conclusions.

II. MODEL AND METHODS

We start from the two-orbital Hubbard model,

$$H = -\sum_{(ij),l} t_{il} C_{il\sigma}^+ C_{jl\sigma} + \sum_{i\sigma} (\epsilon_i - \mu) C_{il\sigma}^+ C_{il\sigma}$$

$$+ \frac{U}{2} \sum_{i\sigma} n_{i\sigma} n_{i\sigma} + J \frac{2}{2} \sum_{i,l,l',\sigma} C_{il\sigma}^+ C_{il'\sigma} C_{il'\sigma} C_{il\sigma}$$

$$+ \frac{U'}{2} \sum_{i,l,l',\sigma} n_{i\sigma} n_{i\sigma} + \frac{J}{2} \sum_{i,l,l',\sigma} C_{il\sigma}^+ C_{il'\sigma} C_{il'\sigma} C_{il\sigma}.$$

(1)

Here index $l$ ($l'$) = $a$ or $b$ refers to the narrow or wide orbital, and $t_{aa}$ ($t_{bb}$) and $t_{ab}$ denote the $NN$ intraorbital and interorbital hopping integrals, respectively; $U$ and $U'$ represent the on-site intraorbital and the interorbital Coulomb repulsions between electrons; and $J$ denotes the Hund’s coupling. The crystal field splitting arising from the structure distortions is defined as $\epsilon_d = \epsilon_a - \epsilon_b$.

In the presence of the finite hopping, we introduce a canonical transformation\cite{20},

$$C_{i\sigma} = u \alpha_{i\sigma} + v \beta_{i\sigma},$$

$$C_{i\sigma} = -v \alpha_{i\sigma} + u \beta_{i\sigma}.$$

Where $u$ and $v$ are the coefficients of the canonical transformation; $\alpha$ and $\beta$ represent fermion annihilation operators of the new effective orbitals, with only intraorbital hopping $t_\alpha = t_{aa} u^2 + t_{bb} v^2 - t_{ab} u v$ and $t_\beta = t_{aa} u^2 + t_{bb} v^2 + t_{ab} u v$, and vanishing interorbital hopping.

As a result, the two-orbital model with interorbital hopping (Eq. 1) is transformed into a new effective two-orbital model without interorbital hopping,

$$H = -t_\alpha \sum_{(ij),\sigma} \alpha_{i\sigma}^+ \alpha_{j\sigma} - t_\beta \sum_{(ij),\sigma} \beta_{i\sigma}^+ \beta_{j\sigma}$$

$$+ (\epsilon_a - \mu) \sum_{i\sigma} (\alpha_{i\sigma}^+ \alpha_{i\sigma} + \beta_{i\sigma}^+ \beta_{i\sigma})$$

$$- \epsilon_d \sum_{i\sigma} (u^2 \alpha_{i\sigma}^+ \alpha_{i\sigma} - u v (\alpha_{i\sigma}^+ \beta_{i\sigma} + \beta_{i\sigma}^+ \alpha_{i\sigma}) + u^2 \beta_{i\sigma}^+ \beta_{i\sigma})$$

$$+ H_{\text{int}}.$$

(2)

Here $H_{\text{int}}$ is the interaction Hamiltonian under the canonical transformation. From Eq. (2), we can distinguish the effects of the interorbital hopping from the crystal-field splitting more clearly within the new fermion representation. That is, the finite interorbital hopping $t_{ab}$ increases the bandwidth ratio of the two effective orbitals; while the crystal field splitting $\epsilon_d$ induces a certain extent of the orbital hybridization of the two effective orbitals at the same site. When $\epsilon_d = 0$, the Green’s function is diagonal in the new fermion representations, and we can employ the DMFT approach to study the two-orbital model easily. On the other hand, when both the crystal field splitting and the interorbital hopping are taken into account, the off-diagonal components of the Green’s function appear in this fermion representation.

In this situation, to discuss the effect of crystal field splitting, we can use the similar technique adopted by Medici et al.\cite{11} and Buenemann et al.\cite{12} for the two-orbital models with the on-site orbital hybridization. In addition, further transformation can be introduced to diagonal the Green’s function. More details of our method are given in Sec. III C.

In the DMFT approach, the self-consistent loop is built to fulfil two equations,

$$G_{\text{imp}}^l(\omega) = G^l(\omega)$$

(3)

$$\Sigma_{\text{imp}}^l(\omega) = \Sigma^l(\omega).$$

(4)

Where $G_{\text{imp}}^l(\omega)$ and $\Sigma_{\text{imp}}^l(\omega)$ represent respectively the Green’s function and the self-energy of the Anderson impurity model mapped from the two-orbital Hubbard model. We adopt the ELDMFT approach\cite{19}, which is a fast and efficient method for the multi-orbital systems. The result of ELDMFT for one-band Hubbard model has been compared with that obtained by the exact diagonalization\cite{21}, and numerical renormalization group\cite{22}, solvers, and a good agreement has been achieved in a rather wide $U$ range.

In the DMFT approach with the exact diagonalization and numerical renormalization group solvers, it has been proved that the lower and higher energy behaviors of the self-energy play the crucial roles. In order to facilitate the calculation and to consider the lower and higher frequency features properly, the ELDMFT approach in-
producetwo self-consistent equations:19,20

\[ V_{ij}^2 = z_i \sum_{j \neq i} t_{ij}^2 \]  

and

\[ n_i' = n_{imp} \]  

Where \( V_i \) is the hybridizing parameter in the Anderson model, \( z_i = 1/(1 - dS^2(0)/d\omega) \) is the quasiparticle weight, and \( n_i' \) \((n_{imp}')\) represents the occupation number of the lattice (impurity) model. In addition, the free-particle DOS is a semicircular type. These two equations correspond to those of the conventional DMFT approach in Eq. 4, respectively, and there is no other extra restriction employed in ELDMFT. The ELDMFT method is able to access the entire parameter region since the Luttinger theorem \( \langle N \rangle = nL \) (\( L \) is the number of lattice size) is exactly fulfilled in the whole range of filling.

In the new fermion representation obtained by the canonical transformation, the optical conductivity of each effective orbital in the paramagnetic system without the interorbital hopping reads21,22

\[ \sigma_\alpha = \pi \int d\omega' d\omega D^\dagger(\epsilon) \rho_\alpha^l(\omega') \rho_\alpha^l(\omega' + \omega) \times \frac{f(\omega') - f(\omega' + \omega)}{\omega}, \]  

where \( D^\dagger(\epsilon) \) and \( \rho_\alpha^l(\omega) \) denote the transition matrix elements and the density of states (DOS) in the new orbital channel \( \alpha \) or \( \beta \), respectively; and \( f(\omega) \) is the Fermi-Dirac distribution function. A small spectra broadening (\( \gamma = 0.1 \)) is introduced for the calculations of DOS. When there is no hopping between the new quasiparticle channels, the total optical conductivity \( \sigma(\omega) \) is the summation of the contributions of two quasiparticle orbitals, \( \sigma_\alpha(\omega) \) and \( \sigma_\beta(\omega) \). In the presence of interorbital hopping, the full optical conductivity for the Green’s functions and the transition matrix \( D^\dagger(\epsilon) \) with off-diagonal terms can be obtained by,

\[ \sigma(\omega) = \pi \int d\omega' d\omega \sum_{l,l'} D_{l'}^\dagger(\epsilon) \rho_l^\dagger(\omega') \rho_l^l(\omega' + \omega) \times \frac{f(\omega') - f(\omega' + \omega)}{\omega}. \]

In this paper, we mainly focus on the optical conductivity of each orbital, \( \sigma_l \), so as to elucidate the metallic or insulating nature of each orbital.

In many transition-metal oxides, the transition-metal ions and oxygen ions usually form TMOn octahedra. The TMO6 clusters distorted from the perfect octahedra contribute the crystal field splitting. Starting from the single-particle potential \( V(\vec{r}) \), one can estimate the splittings in the point-charge approximation of the crystalline field theory.23 Expanding the single-particle potential \( V(\vec{r}) = \sum_j q_j / |\vec{R}_j - \vec{r}| \) in terms of the spherical harmonics:

\[ V(\vec{r}) = \sum_{k=0}^{\infty} \sum_{m=-k}^{k} A_{km} r^k Y_{km}(\theta, \phi) \]

where \( A_{km} = \frac{6}{2k+1} \sum_j \frac{q_j}{|\vec{R}_j|^3} Y_{km}^*(\theta_j, \phi_j) \). The contribution of the crystal field Hamiltonian, \( H_{CF} \), reads

\[ H_{CF} = \sum_\sigma \int d^3r \psi_\sigma^* (\vec{r}) V(\vec{r}) \psi_\sigma (\vec{r}) d\vec{r}. \]

In the second quantization representation, \( H_{CF} \) can be rewritten as

\[ H_{CF} = \sum_j \sum_{\nu \nu'} V_{j\nu} C_{j\nu}^\dagger C_{j\nu'} \]

with \( \psi_\sigma(\vec{r}) = \sum \sum \psi_{j\nu}(\vec{r}) e_{j\nu \sigma} \) and \( \psi_{j\nu}(\vec{r}) \) being the Wannier basis consists of the wavefunctions of the transition-metal d orbits and of the nearest-neighbor atomic orbits. In the high symmetric situation, the matrix \( V_{\nu \nu'} \) is diagonal, giving rise to \( \epsilon_l = V_{\nu \nu'} \). Later, we will evaluate the crystal field splitting based on these equations and the experimental data of the crystal structures of the family compounds Ca2−xSr2RuO4.

III. RESULTS

We study the effects of interorbital hopping and crystal field splitting separately in the asymmetric model, and discuss the application to real compound Ca2−xSr2RuO4. The calculations are performed for the Bethe lattice with semicircular DOS, and our results are measured in units of the hopping integral \( t_b \). In our calculation, the bandwidths of the free-particle spectra are fixed as \( W_a = 2 \) and \( W_b = 4 \).

A. OSMT and optical conductivity

To present the distinction of the optical conductivity in different phases, we first investigate the simplest two-orbital model without interorbital hopping \( (t_{ab} = 0) \) and crystal field splitting \( (\epsilon_d = 0) \). The quasiparticle weight \( Z \) is a powerful tool to characterize the MIT and the OSMT. We present the evolution of quasiparticle weight with the interaction \( U \) in Fig. 1(a) for a two-orbital model with different orbital bandwidths \( W_1/W_2 = 0.5 \). We find that when the Coulomb interaction \( U \) increases to \( U_{c_1} = 1.45 \), the first MIT, i.e. the OSMT, happens. The second transition occurring at \( U_{c_2} = 2.90 \) is the conventional MIT.
The quasiparticle contribution to the optical conductivity arises from the electronic transitions between the lower and the upper Hubbard subbands. As shown in Fig. 1(b), the Drude peaks of both orbitals are considerably large as $U < U_{c1}$, which demonstrates that the system is in the metallic phase. On the contrary, the Drude peaks of both orbitals vanish completely in the insulating region with $U > U_{c2}$, as shown in Fig. 1(d). However, in the OMST regime with $U_{c1} < U < U_{c2}$, the Drude peak of the narrow band vanishes and only the wide band contributes to the conduction channel in the optical conductivity, as shown in Fig. 1(c). Similar to the orbital ordering in manganites and vanadates, the OSMT phase is also orbital polarized in the wide channel. Therefore, the OSMT phase can be detected by using the polarized synchrotron radiation light to measure the distinct optical conduction behaviors of different orbitals.

Apart from the low-energy Drude-like feature coming from the coherent quasiparticle DOS existing near $E_F$, there is another spectral feature, as shown in Fig. 1(b)-(d), centered at the intermediate energy, which represents the charge transfer from the lower and upper Hubbard subbands. Since the general feature of DOS, i.e., the lower and upper Hubbard subbands and a quasiparticle resonance at Fermi surface, can be reproduced by ELDMDFT, our numerical results of optical conductivity are qualitatively accurate. To quantitatively compare with experiments on Ca$_{2-x}$Sr$_x$RuO$_4$, more accurate DOS should be adopted rather than the semicircular DOS, and all three $t_{2g}$ orbitals should be taken into account. This needs to be studied in future work.

### B. Effect of interorbital hopping

Since the interorbital hopping is significant in real material, it is crucial to dig out its effect on the phase diagram and the optical conductivity. To our knowledge, no work has been done to take this important factor into consideration. Till now, only the hybridization of the two orbitals at the same site had been studied by DMFT, where the OSMT tends to be destroyed. However, the physical origins of the on-site intra-atomic hybridization and NN interorbital hopping are quite different. By analyzing the quasiparticle weight at Fermi surface, we obtain the phase diagram of the two-orbital Hubbard model with interorbital hopping integrals $t_{ab} = 0.5$, as shown in Fig. 2(b). For the convenience of comparison, the phase diagram for $t_{ab} = 0.0$ is also shown in Fig. 2(a). In both cases, a stable OSMT regime (the red shadow) lies between the Mott insulating (denoted by "I") and the conventional metallic (denoted by "M") phases. When the interorbital hopping is finite, for example $t_{ab} = 0.5$, the area of the OSMT regime greatly expands, in comparison with the case $t_{ab} = 0$.

It should be noted that the OSMT we observed here is for the effective orbitals in the new fermion representation, which are the hybridized orbitals of the two realistic orbitals. Within this representation, one could find that the bandwidth ratio of the current two effective orbitals increases with the increase of the interorbital hopping integrals $t_{ab}$; the wide quasiparticle band becomes wider, while the narrow one becomes narrower. Thus the OSMT occurs more easily, and its area expands a lot in the phase diagram for large $t_{ab}$. In addition, we also find that for zero and nonzero interorbital hopping, the OSMT regime is close to the $U = U'$ line, but never crosses it. Our result of $t_{ab} = 0$ agrees with Medici et al.'s phase diagram obtained by slave boson approach; however, at $J = 0.5$, our result is slightly different from the result obtained by Koga et al. for $J = 0$ in which the OSMT regime crosses the $U = U'$ line.

The important role of the interorbital hopping $t_{ab}$ in the OSMT also manifests in the optical conductivity. Starting from an insulating case with $U = 2.25$, $U' = 0$ and $t_{ab} = 0$, we obtain the typical feature of insulating optical conductivity for both the narrow band and the
C. Effect of crystal field splitting

The level splitting of the two orbitals \( \epsilon_d \) arising from the crystal field is another important factor that influences the essential of the MIT and the OSMT in the two-orbital systems. To study the evolution of the optical conductivity of the compound \( \text{Ca}_{2-x} \text{Sr}_x \text{RuO}_4 \) with the crystal field splitting \( \epsilon_d \), we start from an OSMT phase with the parameters \( U = 2.0, U' = 1.0 \) and \( J = 0.5 \), which lies in the red shadow regime in the phase diagram shown in Fig. 3(b).

As shown in Eq. (2), there are hybridization terms \( \alpha_{\alpha}^{\alpha} \beta_{\sigma} \) and \( \beta_{\alpha}^{\alpha} \alpha_{\sigma} \) for the two effective orbitals on the same lattice when both the interorbital hopping and crystal field splitting are nonzero. In this condition, the Green’s function is defined as,

\[
G_0(\omega, k)^{-1} = \begin{pmatrix}
\omega + \mu - \epsilon_\alpha(k) & \epsilon_d uv \\
\epsilon_d uv & \omega + \mu - \epsilon_\beta(k)
\end{pmatrix},
\]  

(13)

where \( \epsilon_\alpha(k) \) and \( \epsilon_\beta(k) \) are the bare dispersion relations for the \( \alpha \) and \( \beta \) effective orbitals, respectively. Obviously, we can directly define the local Green function as

\[
G^{\alpha(\beta)}(\omega, x) = \int_{-\infty}^{\infty} dx \rho_0(x) \frac{\rho_0(x)}{\omega + \mu - x - \Sigma^{\alpha(\beta)}(\omega) - \Delta^{\alpha(\beta)}(\omega, x)},
\]  

(14)

Here \( \Sigma^{\alpha(\beta)}(\omega) \) and \( \Sigma^{\alpha(\beta)}(\omega) \) represent the diagonal and off-diagonal terms of the self-energy matrix, respectively.

Based on the special feature of the above off-diagonal Green’s function in Eq. (13), we can introduce an unitary matrix \( U(\omega, k) \) to diagonalize Eq. (13) as an effective Green’s function,

\[
\hat{G}_0(\omega, k)^{-1} = U G_0(\omega, k)^{-1} U^{-1} = \begin{pmatrix}
\omega + \mu - \epsilon^+(k) & 0 \\
0 & \omega + \mu - \epsilon^-(k)
\end{pmatrix},
\]  

(16)

with

\[
\epsilon^{\pm}(k) = \frac{1}{2} (\epsilon_\alpha(k) + \epsilon_\beta(k) \pm \sqrt{(\epsilon_\alpha(k) - \epsilon_\beta(k))^2 + 4\epsilon_d^2 u^2 v^2}).
\]  

(17)

For a half-filling asymmetric two-orbital system, when the relationship \( |\epsilon_\alpha(k) - \epsilon_\beta(k)| < |\epsilon_d| \) is satisfied in the energy region close to the Fermi surface, we can approximate the above dispersion relations in the first order as,

\[
\epsilon^{\pm}(k) = \frac{1}{2} (\epsilon_\alpha(k) + \epsilon_\beta(k) \pm (2|\epsilon_d u| + (\epsilon_\alpha(k) - \epsilon_\beta(k))^2 / 4|\epsilon_d u|)).
\]  

(18)

It is obvious that the finite crystal field splitting mixes the two effective \( \alpha \)- and \( \beta \)-orbital in the low energy regime, and as a result, the OSMT is suppressed.

We employ the ELDMFT approach to numerically solve the corresponding impurity problem, and find that the OSMT phase is strongly influenced by the transfer of electrons between the two effective orbitals. We start from an OSMT phase with \( \epsilon_d = 0.0 \), in which the electrons in the narrow orbital are localized and itinerant in the wide one, respectively. As shown in Fig. 3(c), the OSMT character clearly manifests in the Drude peak of
each orbital: the Drude peak vanishes completely for insulating narrow orbital, while the wide orbital is metallic with a finite Drude peak.

When the crystal field splitting becomes negative as \( \epsilon_d = -0.5 \) (see Fig. 4(a)) and \(-0.25\) (see Fig. 4(b)), the narrow orbital lifts to the lower energy region, and a fraction of electrons in the wide orbital transfers to the narrow orbital. The electron filling in the narrow orbital is more than a half, thus the narrow orbital also becomes metallic. In this situation, the Drude peak of the narrow orbital appears again. On the other hand, as the crystal field splitting is positive, the narrow orbital keeps its insulating character and the wide one is metallic. So the system remains in the OSMT phase, as shown in Fig. 4(d).

Furthermore, we find that in both the metallic and insulating phases near the border of the OSMT regime, a small negative crystal field splitting can drive neither the metallic system nor the insulating system into the OSMT phase. Therefore, the negative crystal field splitting is dominant. Noting that though the single-ion crystal field theory is relatively simple, it is qualitatively correct for the sign and the relative magnitude of the crystal field splittings. As shown in Fig. 5(a), the averaged splittings between the \( d_{xz/yz} \) and the \( d_{xy} \) bands are \( \epsilon_d/\epsilon_{d0} = -0.013, -0.11, -0.012, 0.0025 \) and 0.005 for \( x=2.0, 0.5, 0.2, 0.1 \) and 0, respectively. Here \( \epsilon_{d0} \) is a positive constant, representing the 4d-orbital radial integral of the single-particle potential \( (\epsilon_{d0} = \int_0^\infty r^2dr\langle\phi(r)\rangle V(r)\langle\phi(r)\rangle) \). The level separation of the narrow \( d_{xz/yz} \) bands from the wide \( d_{xy} \) band in Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) varies gradually from positive to negative with the increase of Sr concentration, and have the largest value as \( x = 0.5 \). This largest level separation arises from the rotation-type distortion \( 26 \), which becomes the most significant when \( x = 0.5 \). As \( x \) further decreases, the tilting distortion of the RuO\(_6\) octahedra \( 25 \) becomes more important, which usually mixes different orbitals.

Since the normal state of SrRuO\(_4\) is metallic, our preceding results show that negative crystal field splitting is unfavorable of stable OSMT phase. Therefore, our results exclude the possibility of the OSMT occurring in Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) when \( x \) increases from 0.2 to 2.0. Very recently, Liebsch \textit{et al.} \textsuperscript{28} suggested that in compound Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\), the splitting of the \( d_{xz/yz} \) to the \( d_{xy} \) bands is positive. In their study, the band structures in compound Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) is rigid, and thus the variation of electron occupations in different orbitals arises from the change of the positive crystal field splitting. Based on this hypothesis, Liebsch \textit{et al.} \textsuperscript{28} and Werner \textit{et al.} \textsuperscript{27} discussed the role of the positive crystal field splitting on the MIT in two-orbital Hubbard model. However, Lee \textit{et al.} \textsuperscript{22} have demonstrated that with the doping increasing from \( x=0 \) to 2, the bandwidths of the \( d_{xy} \) and \( d_{xz/yz} \) orbitals considerably change, implying that the rigid band approximation is not proper.

**D. Application to Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\)**

For the compound Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\), the crystal field splitting arises from the structure distortions, and it can be approximately modeled as a two-orbital system since the double degeneracy of the narrow \( d_{xz/yz} \) bands is kept over the whole doping range. Within the framework of the single-ion crystal field theory\textsuperscript{24}, we investigate the crystal field splitting between the \( d_{xz/yz} \) orbitals and the \( d_{xy} \) orbital with the doping concentration \( x \) varying from 0.0 to 2.0. According to the crystal structures for \( x=2.0, 0.5, 0.2, 0.1 \) and 0\textsuperscript{22}, we find that in the most doping region, the narrow \( d_{xz/yz} \) bands are lower than the broad \( d_{xy} \) band, showing that the negative crystal field splitting is dominant. Noting that though the single-ion crystal field theory is relatively simple, it is qualitatively correct for the sign and the relative magnitude of the crystal field splittings. As shown in Fig. 5(a), the averaged splittings between the \( d_{xz/yz} \) and the \( d_{xy} \) bands are \( \epsilon_d/\epsilon_{d0} = -0.013, -0.11, -0.012, 0.0025 \) and 0.005 for \( x=2.0, 0.5, 0.2, 0.1 \) and 0, respectively. Here \( \epsilon_{d0} \) is a positive constant, representing the 4d-orbital radial integral of the single-particle potential \( (\epsilon_{d0} = \int_0^\infty r^2dr\langle\phi(r)\rangle V(r)\langle\phi(r)\rangle) \). The level separation of the narrow \( d_{xz/yz} \) bands from the wide \( d_{xy} \) band in Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) varies gradually from positive to negative with the increase of Sr concentration, and have the largest value as \( x = 0.5 \). This largest level separation arises from the rotation-type distortion \( 26 \), which becomes the most significant when \( x = 0.5 \). As \( x \) further decreases, the tilting distortion of the RuO\(_6\) octahedra \( 25 \) becomes more important, which usually mixes different orbitals.

Since the normal state of SrRuO\(_4\) is metallic, our preceding results show that negative crystal field splitting is unfavorable of stable OSMT phase. Therefore, our results exclude the possibility of the OSMT occurring in Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) when \( x \) increases from 0.2 to 2.0. Very recently, Liebsch \textit{et al.} \textsuperscript{28} suggested that in compound Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\), the splitting of the \( d_{xz/yz} \) to the \( d_{xy} \) bands is positive. In their study, the band structures in compound Ca\(_{2-x}\)Sr\(_x\)RuO\(_4\) is rigid, and thus the variation of electron occupations in different orbitals arises from the change of the positive crystal field splitting. Based on this hypothesis, Liebsch \textit{et al.} \textsuperscript{28} and Werner \textit{et al.} \textsuperscript{27} discussed the role of the positive crystal field splitting on the MIT in two-orbital Hubbard model. However, Lee \textit{et al.} \textsuperscript{22} have demonstrated that with the doping increasing from \( x=0 \) to 2, the bandwidths of the \( d_{xy} \) and \( d_{xz/yz} \) orbitals considerably change, implying that the rigid band approximation is not proper.
Based on the above findings, the two successive MIT in Ca$_{2-x}$Sr$_x$Ru$_4$O$_{12}$ is obviously independent of the OSMT. A question arises: what happens in the first unconventional MIT at $x=0.2$? We notice that at the doping concentration of $x=0$ and $x=0.5$, the doped ruthenates are found to be orbital ordering. Therefore, the two successive MIT should be accompanied by the transitions of the ordered orbital states, which have not been well treated in the literature. To account for the properties of these two MIT, the orbital ordering should be taken into account properly within the DMFT approach, which has gone beyond the scope of this paper.

IV. CONCLUSIONS

We have studied the effects of two important factors, i.e. the crystal field splitting and interorbital hopping, on OSMT in nondegenerate two-orbital models and in the compounds Ca$_{2-x}$Sr$_x$Ru$_4$O$_{12}$. Our results show that the negative crystal field splitting can hybridize the two non-degenerate orbitals at the same lattice site, and strongly restrain the appearance of the OSMT phase. However, the finite interorbital hopping considerably expands the OSMT region in the phase diagram. In the doping region form $x = 0.2$ to 2.0 in Ca$_{2-x}$Sr$_x$Ru$_4$O$_{12}$, the crystal field splitting introduced by the distortion of the RuO$_6$ octahedra is found to be negative. Therefore, the occurrence of OSMT in compound Ca$_{2-x}$Sr$_x$Ru$_4$O$_{12}$ unlikely happens in the corresponding doping region.

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