

Lifshitz-Transition-Driven Metal-Insulator Transition in Moderately Spin-Orbit-Coupled $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$

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 (Received 21 November 2018; revised manuscript received 31 May 2019; published 4 September 2019)

Motivated by the novel insulating state of Sr_2IrO_4 from strong spin-orbit coupling (SOC), we investigate, by means of angle resolved photoemission, the metal-insulator transition (MIT) mechanism in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ whose mother compound is isovalent and isostructural but has smaller SOC strength compared to Sr_2IrO_4 . Transport and angle resolved photoemission results from single crystalline $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ revealed that the MIT occurs coincidentally with a multi- to single-band transition (Lifshitz transition) at $x = 0.4$. Starting from $x = 0.4$, there is a gradual but anomalous enhancement in the band gap size with additional electron doping, suggesting that the insulating phase in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is a new type which has been rarely investigated. These results suggest that the insulating phase in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is likely induced by the moderate SOC strength and electron doping effect from the La. Our findings not only elucidate the MIT mechanism in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$, but may also open new avenues for novel MIT research in moderate SOC regimes.

DOI: [10.1103/PhysRevLett.123.106401](https://doi.org/10.1103/PhysRevLett.123.106401)

Mott physics has been one of the most intensively studied subjects in condensed matter physics. In early studies, the main focus was on materials with large on-site Coulomb repulsion (U) in localized orbitals such as $3d$ transition metal oxides (TMOs). However, discovery of the relativistic Mott insulating state in Sr_2IrO_4 [1] has led to extensive studies on $5d$ TMOs with an expectation that exotic phenomena due to strong spin-orbit coupling (SOC) may exist. In Sr_2IrO_4 , the large SOC transforms the usual crystal field split eigenstates into J_{eff} states, and then the narrow bandwidth (W) of the $J_{\text{eff}} = 1/2$ states which is comparable to U results in a relativistic Mott state, forming upper and lower Hubbard bands. The insulating state is found to be easily broken with a small amount of charge carrier doping [2] as observed in $\text{Sr}_{2-x}\text{La}_x\text{IrO}_4$, which indicates the fragile nature of the Mott state against carrier doping.

On the other hand, SOC in the $4d$ TMO is smaller and is thus expected to play, in comparison to Sr_2IrO_4 , a lesser role in the determination of the physical properties. A recent electronic structure study on $\text{Sr}_2\text{Rh}_x\text{Ir}_{1-x}\text{O}_4$ revealed that Rh ($4d$) substitution considerably reduces the SOC strength and leads to the collapse of the relativistic Mott state in Sr_2IrO_4 [3]. However, a theoretical study suggests that the electronic structure of Sr_2RhO_4 still can be understood within the J_{eff} scheme even though the SOC strength in Sr_2RhO_4 is much smaller than the case of Sr_2IrO_4 [4]. This indicates that the moderate SOC in $4d$ TMOs may still significantly affect the physical properties

and that an investigation of the role of SOC in $4d$ TMOs may lead us to new exotic phases as in the case of Sr_2IrO_4 .

In that regard, we focus our attention on the anomalous metal-insulator transition (MIT) in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. The resistivity of $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ gradually increases with the La ($5d6s^2$) substitution for Sr ($5s^2$) [5], turning the system from a metal into an insulating phase. This result is opposite to what is theoretically predicted in which itinerant metallic Stoner-type ferromagnetism is induced upon La doping to Sr_2RhO_4 [6], thus the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ has been understood within the Anderson localization picture from the disorder effect of La dopants [5,7]. However, our experimental results are not consistent with such interpretation and thus the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is yet to be fully understood.

In this Letter, we report our electronic structure data regarding the exotic MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Our result shows a clear emergence of band gap through the MIT, which is found to be inconsistent with previous assertions of MIT through the Anderson localization [5,7]. In addition, the gradual increase of the band gap in the noninteger electron filling region starting from $x = 0.4$ suggests the insulating state in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is totally different from what has been investigated so far. We find that the exotic MIT may be attributed to the interplay between moderate SOC and U with the help of the electron doping effect from La substitution. Our result not only is a discovery of a new type of insulating state in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ but also

introduces a new mechanism for novel MIT in which moderate SOC plays a significant role.

The $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ single crystals were grown from off-stoichiometric quantities of SrCO_3 , Rh_2O_3 , and La_2O_3 in order to compensate volatility of Rh_2O_3 during the growth using the floating zone method. The powder was thoroughly ground and calcined in flowing O_2 . Calcined polycrystalline powder was then pressed into rods and sintered at 1300°C in flowing O_2 gas for 4 h. The crystal growth was performed in an image furnace with 10 atm O_2 pressure. The growth speed was 10 mm/h. To compensate oxygen deficiencies due to the multivalence nature of Rh, all samples were annealed at 1100°C in flowing O_2 gas after the growth. Resistivity measurement was done with the Quantum Design physical property measurement system using the four-probe method. ARPES measurements were performed at the beam line 4.0.3 of the Advanced Light Source, Lawrence Berkeley National Laboratory. Samples were cleaved *in situ* and measurements were performed with $h\nu = 70$ eV at 30 K in an ultrahigh vacuum better than 5×10^{-11} Torr.

Doping dependent electronic structure results are shown in Fig. 1. Two original as well as folded Fermi surface pockets of Sr_2RhO_4 are seen in Fig. 1(a), which is consistent with previous ARPES studies [8]. The larger Fermi surface is usually labeled as β while the smaller pocket as α . The energy dispersion in Fig. 1(h) indicates that the holelike α band (green dashed line) is occupied more than the β band (yellow dashed line). According to a

theoretical study [4], the α and β bands approximately correspond to $J_{\text{eff}} = 3/2$ with $|m_{J_{\text{eff}}}| = 3/2$ and $J_{\text{eff}} = 1/2$ bands, respectively. Over the entire range of La content, the α band shows a rigid-band-like shift due to the electron doping from La. The α pocket size decreases until $x = 0.4$ [Figs. 1(a)–1(d) and 1(h)–1(k)]. Then the top of α band gradually sinks below the Fermi level for $x > 0.4$ [Figs. 1(e)–1(g) and 1(l)–1(n)]. On the other hand, the Fermi surface size of the β band increases with the La content until $x = 0.4$ [Figs. 1(a)–1(d), 1(h)–1(k)], and then it fades out from the Fermi level with a gradually increasing band gap [Figs. 1(e)–1(g), 1(l)–1(n)]. These observations indicate that the critical doping level for the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is $x = 0.4$. This value for the critical doping is also consistent with that obtained from the result of resistivity measurements on single crystalline $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ (Supplemental Material, Fig. S1 [9]).

The band gap evolution as a function of the La content is easily seen in symmetrized energy distribution curves (EDCs) plotted in Figs. 2(a) and 2(b). The plotted EDCs are obtained by averaging the EDCs from the regions near α and β bands, respectively [shaded in blue in Fig. 1(a)]. In order to investigate the evolution of the band gap systematically, we plot the band top energy as a function of La concentration in Fig. 2(c). The band top energy is defined as the x -axis intercept value of the linear fit of the leading edge (T_α and T_β for α and β bands, respectively). The resulting doping dependent T_α and T_β are plotted in Fig. 2(c). As can be seen in Fig. 2(c), both T_α and T_β are

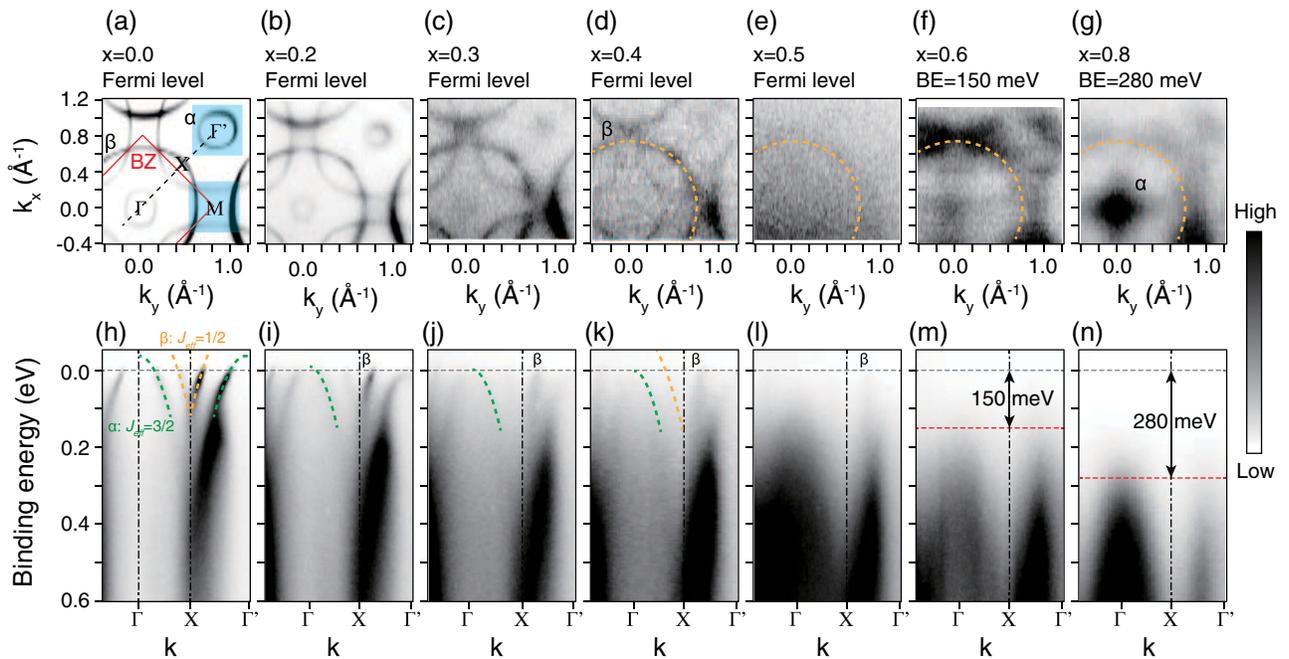


FIG. 1. ARPES data from $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. (a)–(g) Constant binding energy $k_x - k_y$ maps at the binding energies of 0 meV ($x = 0.0$, 0.2, 0.3, 0.4 and 0.5), 150 meV ($x = 0.6$), and 280 meV ($x = 0.8$). The squares shaded in blue in panel (a) are integration regions for α and β band spectra in Figs. 2(a) and 2(b). (h)–(n) ARPES data along the Γ -X line. The band gap size for $x = 0.6$ (m) and $x = 0.8$ (n) is also shown.

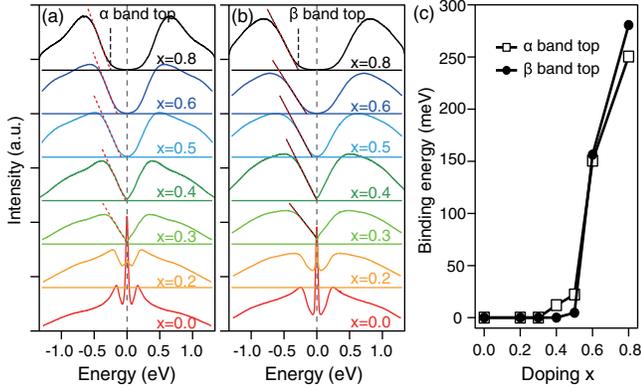


FIG. 2. (a),(b) Momentum averaged energy distribution curves (EDCs) from the regions of (a) α and (b) β bands [blue shaded squares in Fig. 1(a)]. (c) Doping dependent band top energies for α (T_α) and β (T_β) bands. T_α and T_β are defined as the x -axis intercept value of a linear fit to the leading edge.

zero for $x < 0.4$, which indicates α and β bands cross the Fermi level, and then monotonically increase from $x = 0.4$. It is worth noting that T_α and T_β show almost identical behaviors over the entire doping levels. Such synchronized behavior is unexpected since the two bands are independent and well separated in the energy-momentum space as shown in Fig. 1(h).

Before we proceed to the details of the doping dependent electronic structure, we briefly discuss possible scenarios for the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Our results clearly show that the MIT appears at $x = 0.4$, which means that the total number of electrons in the Rh $4d$ orbitals is about 5.4 considering each La substitution provides an extra electron. Therefore, standard theories for band and Mott insulators cannot account for the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ as the electron filling is a noninteger. An electronic or magnetic order might trigger a transition to an insulating phase. However, our measurements (Supplemental Material, Fig. S1 [9]) as well as those previously reported [5,7] show that there is no signature for such ordered phases in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Another candidate we can consider is a structural transition from tetragonal to orthorhombic which originates from the emergence of out-of-plane octahedral tilting distortion along the [110] direction [5,7]. This transition can also be counted out as the cause of the MIT since the octahedral out-of-plane distortion along the [110] axis does not reduce the crystal symmetry that is needed to generate a hybridization gap [5,15]. All of these point to a situation that the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ is an unexpected phenomenon which cannot be easily explained. For this reason, the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ has been attributed to a disorder effect-driven phenomenon (Anderson localization) in previous reports [5,7]. However, our ARPES result clearly contradicts such an interpretation as it shows a fairly well-defined hard gap in the electronic structure that is unexpected for Anderson localization

insulators [16,17]. In addition, our temperature dependent resistivity measurements also suggest that Anderson localization is not an appropriate model to explain the MIT (Supplemental Material, Fig. S1 [9]).

The proceeding discussion suggests that we need a different mechanism to explain the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. To unravel the MIT mechanism in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$, we may need further information from the measured electronic structure of $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Since La substitutes Sr (not Rh which determines the near Fermi energy electronic structure), we can assume that the $4d$ orbital character remains intact upon the La substitution. Indeed, as mentioned above [Figs. 1(a)–1(c) and 1(h)–1(j)], there is only an approximate downward rigid-band-like shift upon La substitution without noticeable electronic structure change, up to $x = 0.4$. It is thus reasonable to assume that the main effect of La substitution is simple electron doping. It is interesting to note that, as La content increases, the insulating phase appears exactly when the α band becomes fully occupied at $x = 0.4$. This observation makes us suspect that a full occupation of the α band may trigger the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. That is, the multiband to single-band transition (or Lifshitz transition) coming from the full occupation of the α band might be the key factor in the MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$.

The first step is to see if the localized character of electrons, an essential ingredient of correlated insulators, is enhanced across $x = 0.4$. In that respect, the single-band nature of the band should be more advantageous than the multiband state. The statement can be intuitively understood in terms of conduction bandwidth as schematically illustrated in Fig. 3. In the multiband regime ($x < 0.4$), partially filled α and β bands are involved in the electron transport, and both bands should be considered for the bandwidth W of the conduction electrons (see the left two panels in Fig. 3). On the other hand, only the electrons in the β band can move in the single-band regime ($x > 0.4$), and the effective bandwidth thus becomes significantly reduced compared to the multiband regime (see the right two panels in Fig. 3). In addition, electron doping is found to further reduce the bandwidth of the β band. In a previous study on Sr_2RhO_4 , the concept of “effective SOC strength”

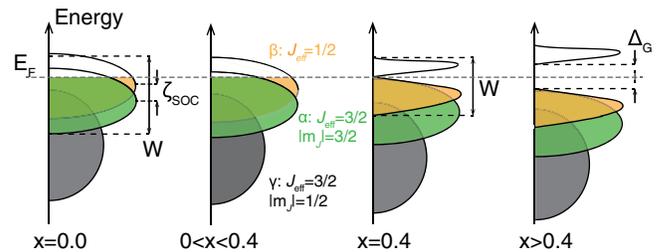


FIG. 3. Schematic illustration of the evolution of the electronic density of states with La substitution. Multiband ($x < 0.4$) to single-band ($x > 0.4$) transition by electron (La) doping reduces the bandwidth W . In the single-band region ($x > 0.4$), a band gap (Δ_G) develops.

has been proposed to explain the experimentally observed large SOC effect compared to what is expected from the atomic SOC strength of Rh [18]. Our theoretical study (Supplemental Material, Sec. V [9]) reveals that electron doping in Sr_2RhO_4 leads to a smaller effective SOC strength which in turn results in a reduction of the β bandwidth by mixing constituent J_{eff} states. In brief, Lifshitz transition should be the main culprit for the enhanced electron localization in the β band while the reduced bandwidth from the decrease in the effective SOC upon electron doping is an additory factor.

The synchronized behavior between T_α and T_β in Fig. 2(c) can be well understood within the picture depicted in Fig. 3. As the electron doping increases, the Fermi energy E_F eventually rises above the top of the α band, with the T_α being the difference between E_F and the α band top. Then, the system is in the single-band regime with the β band and a correlation gap T_β develops. However, such a single-band regime is limited to the energy region between E_F and the α band top. Therefore, the gap in the β band (T_β) cannot exceed T_α (Fig. 3) because α and β bands energetically overlap below the α band top, allowing interband interactions which are detrimental to electron localization. These observations lead to the synchronized behavior of T_α and T_β . It is worth noting that, contrary to the fragile nature of the usual Mott insulating state against charge carrier doping, the insulating state of $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ shows a gradual enhancement of band gap size with electron doping [Figs. 2(a) and 2(b)]. This, in combination with its non-integer electron filling, strongly implies a new type of insulating state which has not been studied yet.

We attribute the emergence of a gradually increasing band gap in the insulating state to the moderate SOC in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Here, we briefly discuss the role of SOC in the formation of such an insulating state. Strong SOC is an essential ingredient of the Mott state in $5d$ Sr_2IrO_4 (~ 0.4 eV) since it splits t_{2g} bands into narrower J_{eff} states, $J_{\text{eff}} = 3/2$ and $J_{\text{eff}} = 1/2$. Meanwhile, the SOC in Sr_2RhO_4 (~ 0.15 eV) is not strong enough to induce fully occupied $J_{\text{eff}} = 3/2$ and half-filled $J_{\text{eff}} = 1/2$, and this is the reason why Sr_2RhO_4 is a metal (Supplemental Material, Figs. S3 and S4 [9]). However, we believe the moderate SOC in Sr_2RhO_4 plays the key role in the novel insulating behavior. First of all, the SOC in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ can still lead to narrow and parallel bands required for a relativistic Mott insulating state. In addition, a moderate SOC is an essential condition to have the unique insulating state in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ —the gradually increasing band gap with the La concentration. As we explained above, T_α is the key parameter that determines T_β . Therefore, a necessary condition for the band gap variation is that the α band should be located near the Fermi level. Noting that the SOC strength is the key parameter to determine splitting size between α and β bands (Supplemental Material, Fig. S4 [9]), a moderate SOC is indeed an essential

ingredient of the novel insulating phase. In brief, moderate SOC is the only way we could find to generate a gradually increasing band gap behavior since it provides SOC strong enough to induce the eventual correlated insulating gap but weak enough to have incomplete splitting between J_{eff} states.

A remaining question is if there is an effective model that can describe the insulating state of $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. As already mentioned, the Mott-Hubbard model (Mott insulator) cannot account for the insulating state in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ which has a noninteger electron filling. Furthermore, the observed hard band gap in the electronic structure (Fig. 2) suggests that the disorder-driven electron localization scenario (i.e., Anderson localization) is not the one either [16,17]. Instead, a possible candidate may be the Anderson-Hubbard (AH) model [19] which covers strong electron correlation as well as disorder. The AH Hamiltonian is given as [19]

$$H = -t \sum_{\langle i,j \rangle \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_{i\sigma} W_i n_{i\sigma}, \quad (1)$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) is the creation (annihilation) operator for electron at site i , σ the spin, t the hopping between the nearest-neighbor sites, $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ a local electron number operator at site i , U the on-site Coulomb repulsion energy, and W_i a random potential energy at each site. The first two terms are from the Mott-Hubbard Hamiltonian and the last term accounts for the disorder contribution. One of the most important aspects of the AH Hamiltonian is its capability to describe noninteger filling Mott insulating states by considering the effect [the third term in Eq. (2)]. Several studies show that impurities in the system make the Mott state robust against charge carrier doping [20–23]. In this respect, AH is a model that may provide a theoretical starting point to account for the noninteger filling insulating states in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$. Therefore, we expect that the AH Hamiltonian can be a possible candidate to fully account for MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ if the role of the moderate SOC effect can be additionally considered.

Our studies on MIT in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ have important implications. The first is on the discovery of the exotic insulating state. While ordinary insulating states are quite fragile against charge carrier doping, the insulating state in $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ ($x > 0.4$) is robust and has a gap size that is approximately proportional to the electron doping. This type of insulator has not been investigated and it certainly defies the conventional knowledge on Mott insulators. Therefore, our study can be further expanded to explore new types of insulators like $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$.

While there are recent discoveries of exotic phenomena, such as superconductivity, magnetism, and MIT in $4d$ TMOs [24–27], the detailed role of SOC in these compounds has not been clearly understood so far. Our study on $\text{Sr}_{2-x}\text{La}_x\text{RhO}_4$ reveals how SOC can generate unexpected

phenomena by modifying t_{2g} bands, and is a testimony to the important role of SOC even in $4d$ TMOs. Along with the recent attempts to understand the intermediate eigenstates in $4d$ TMOs such as Van-Vleck magnetism in Ca_2RuO_4 [28], we believe our study should initiate further studies that will elucidate various exotic phenomena in systems with moderate SOC.

The authors acknowledge fruitful discussion with T. W. Noh and J. W. Rhim. This work was supported by the Institute for Basic Science in Korea (Grant No. IBS-R009-G2). The Advanced Light Source is supported by the Office of Basic Energy Sciences of the U.S. DOE under Contract No. DE-AC02-05CH11231.

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