Orbital-Dependent Phase Control in Ca$_{2-x}$Sr$_x$RuO$_4$ ($0 \leq x \leq 0.5$)

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Abstract

We present first-principles studies on the orbital states of the layered perovskites Ca$_{2-x}$Sr$_x$RuO$_4$. The crossover from antiferromagnetic (AF) Mott insulator for $x < 0.2$ to nearly ferromagnetic (FM) metal at $x = 0.5$ is characterized by the systematic change of the $xy$ orbital occupation. For the AF side ($x < 0.2$), we present firm evidence for the $xy$ ferro-orbital ordering. It is found that the degeneracy of $t_{2g}$ (or $e_g$) states is lifted robustly due to the two-dimensional (2D) crystal-structure, even without the Jahn-Teller distortion of RuO$_6$. This effect dominates, and the cooperative occupation of $xy$ orbital is concluded. In contrast to recent proposals, the resulting electronic structure explains well both the observed X-ray absorption spectra and the double peak structure of optical conductivity. For the FM side ($x = 0.5$), however, the $xy$ orbital with half filling opens a pseudo-gap in the FM state and contributes to the spin $S=1/2$ moment (rather than $S=1$ for $x=0.0$ case) dominantly, while $yz, zx$ states are itinerant with very small spin polarization, explaining the recent neutron data consistently.

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I. INTRODUCTION

The spin and orbital structures in Ca\textsubscript{2-x}Sr\textsubscript{x}RuO\textsubscript{4} (0 \leq x \leq 0.5) attract much attention recently as a model system of $t_{2g}$ electrons. It is an antiferromagnetic (AF) Mott insulator for $x < 0.2$, but nearly ferromagnetic (FM) metal at $x = 0.5^{1,2,3}$. There are four electrons occupying $t_{2g}$ states per Ru$^{4+}$, while $e_g$ states are empty. The isovalent substitution of Sr for Ca does not change the number of electrons, but modifies the crystal structure systematically\textsuperscript{3} due to their different ionic radii. The key issue here is the orbital degree of freedom, which couples with lattice and magnetism strongly. Extensive experimental\textsuperscript{4,5,6,7,8} and theoretical\textsuperscript{9,10,11,12} studies have been done for this issue, while the present understanding of this compound is quite controversial.

First, for the AF side ($x=0.0$), the preferential occupation of the $xy$ orbital was predicted by earlier calculations\textsuperscript{9,10}. However the O 1s X-ray absorption spectroscopy (XAS) study reported the occupation of about 0.5 holes in the $xy$ orbital\textsuperscript{4}. In addition to this, the photoemission experiment suggested strong spin-orbit coupling (SOC), leading to the proposal of complex $t_{2g}$ orbitals\textsuperscript{4}. The presence of 0.5 holes in the $xy$ orbital also led Hotta et al.’s\textsuperscript{12} to propose the “antiferro-orbital ordering” (AFO) with 2x2 periodicity of RuO\textsubscript{6} in the plane. The recent experiment on optical conductivity was interpreted in terms of this orbital pattern\textsuperscript{5}. However, the lattice distortion corresponding to the AFO (i.e., the 2x2 structure) is not observed experimentally, and the orbital pattern of AFO is not consistent with the recent resonant X-ray scattering (RXS) experiment\textsuperscript{6}, in which no superlattice peak was observed. Second, for the nearly FM side ($x=0.5$), the fitting of the Curie-Weiss type susceptibility suggested $S=1/2$ spin moment\textsuperscript{2}, in sharp contrast with the high-spin ($S=1$) state for $x=0.0$. A similar picture may hold also for $x=0.2^{7}$. The recent polarized neutron diffraction studies for $x=0.5^{8}$ suggested dominant magnetization distribution on the $xy$ orbital rather than the theoretically suggested $yz/zx$ orbitals\textsuperscript{10}. Therefore it is an important and challenging issue to determine the orbital state of Ca\textsubscript{2-x}Sr\textsubscript{x}RuO\textsubscript{4} explaining consistently the available experiments, which we undertake in this paper.

We show that the crossover of magnetic properties with increasing doping $x$ is characterized by the systematic change in the $xy$ orbital occupation. For the AF side ($x < 0.2$), we conclude that the predicted ferro-orbital (FO) ordering with dominant $xy$ occupation explains well (a) the XAS spectra, and (b) the double peak structure of the optical conduc-
tivity. We further point out that the stabilization of the \textit{xy} state is quite robust, and mostly due to the two-dimensional (2D) crystal-field, which has not been considered seriously so far. In contrast to the AF side, for \( x=0.5 \), the electronic structure is characterized with the \( yz/zx \) states with small moment, and the \textit{xy} orbital with half filling, which opens a pseudo-gap in the FM state and contributes to the \( S=1/2 \) moment dominantly. This picture, which is in strong contradiction to the previous proposal \cite{10}, explains the neutron data \cite{8} consistently.

II. \textsc{Calculation Method}

The calculations were done with the first-principles plane-wave pseudopotential method. In our previous work \cite{9}, the effects of structural distortions were emphasized by taking into account important distortions mostly, however here we use the experimental structures to compare the calculated results with the measured spectra. Five experimental structures are considered, i.e., \( x=0.0 \) at 11K and 295K, \( x=0.1, 0.2 \) and \( 0.5 \) at 10K \cite{3}. Two kinds of magnetic states, FM and staggered AF states, are considered for each structure. In the previous work \cite{9}, the local spin density approximation (LSDA) was adopted for the entire range. The validity of LSDA for predicting the stable magnetic state of ruthenates was demonstrated \cite{13}. However for Ca\(_2\)RuO\(_4\), though LSDA can predict the stability of AF magnetic order, the band gap does not open \cite{9,11}. As we aim to analyze the optical conductivity also, a proper reproduction of the band gap is important. Therefore, we employ the LDA+U scheme \cite{14,15} for the insulating region \((0 \leq x < 0.2)\) with \( U_{\text{eff}}=2.5\text{eV} \) to reproduce the band gap of Ca\(_2\)RuO\(_4\). The LSDA was employed for other region of \( x \). The details of other aspects of the calculation were described in the previous paper \cite{9}. For the studies of XAS spectra and optical conductivity, the interband transitions are calculated from the converged Kohn-Sham wave functions and eigen values, by using the core compensation form of pseudo-wave functions (see Ref. \cite{16} for technical details).

III. \textsc{Results and Discussion}

A. \textbf{Ca\(_2\)RuO\(_4\) \((x=0)\)}

We start with Ca\(_2\)RuO\(_4\) at 10K. Our calculations predict the AF ground state with energy gain about 28.2meV with respect to the FM state. The mechanism for stabilizing
the AF state was analyzed in our previous paper in terms of lattice distortion, flattening, rotation and tilting of RuO$_6$. The projected density of states (PDOS) in Fig.1(a) suggests that the present situation may correspond to the localized spin picture of $S=1$, where the $xy$ states are fully occupied and the $yz/zx$ orbitals are half filled. In reality, as the Ru 4$d$ orbitals are extended, inter-site hybridization modifies the picture quantitatively. For example, the occupation numbers (in LDA+U) are given as follows: $n_{xy}^{\uparrow}=0.86$, $n_{yz/zx}^{\uparrow}=0.87$, $n_{xy}^{\downarrow}=0.79$ and $n_{yz/zx}^{\downarrow}=0.28$, with up and down arrows indicating the spins (see Figs.1(a) and 2(a)). The cooperative occupation of the $xy$ orbital for all Ru sites forms the FO ordering. These occupation numbers give 1.25$\mu_B$ as the magnetic moment of a Ru atom $M_{\text{Ru, total}}$, which agrees well with the experimental value of 1.3$\mu_B$. As for the spin polarization of oxygen, the in-plane oxygen atoms have no net polarization due to symmetry while the apical oxygen atoms are strongly polarized (about 0.1 $\mu_B$) due to the strong $p - d$ hybridization because the $yz/zx$ orbitals which extend toward apical oxygen contribute dominantly to the Ru spin polarization. The corresponding values in LSDA for the above orbital population are $n_{xy}^{\uparrow}=0.80$, $n_{yz/zx}^{\uparrow}=0.82$, $n_{xy}^{\downarrow}=0.71$ and $n_{yz/zx}^{\downarrow}=0.40$. Therefore, the basic feature of the orbital population remains the same in LSDA, though the orbital polarization and the spin polarization are reduced compared with those in LDA+U. For example, LSDA gives 0.93$\mu_B$ as $M_{\text{Ru, total}}$. The reduction in these polarizations is due to the stronger inter-site hybridization caused by the vanishing band gap in LSDA. Therefore, we believe that the results obtained by LDA+U which adjusts the effective Coulomb repulsion $U_{\text{eff}}$ to reproduce the band gap should be quantitatively more reliable than those by LSDA. The present results for the electronic structure are consistent with previous calculations.

As for the orbital character, there are three factors mainly contributing to the stabilization of the $xy$ orbital:

1) The energy level splitting due to 2D crystal field in the layered structures (in contrast to its three-dimensional (3D) counterpart, where the $t_{2g}$ (or $e_g$) states are degenerate). Fig.3 (main panel) shows the calculated PDOS for the non-magnetic (NM) state of Ca$_2$RuO$_4$ without the Jahn-Teller distortion (i.e. three Ru-O bonds having equal length). In the inset of Fig.3, we show the calculated energy difference between the $xy$ and the $yz, zx$ band as a function of the tetragonal JT distortion, by calculating the center of gravity for each band. Clearly, even for the hypothetical tetragonal Ca$_2$RuO$_4$ without JT distortion, the $xy$ orbital is lower in energy than the $yz$ and $zx$ orbitals by about 0.2eV as estimated by our
calculation. This lowering of $xy$ state energy is added to that due to the tetragonal distortion as demonstrated in the almost uniform downward shift of the dashed line (Ca$_2$RuO$_4$) from the solid line (CaRuO$_3$) in the inset of Fig.3. As a first approximation, the origin of the energy level splitting is attributed to the following geometrical aspect in the second neighbor configurations: in Ca$_2$RuO$_4$, Ru-O-Ru in the $ab$ plane is replaced with Ru-O-Ca along the $c$ axis. Two factors coming from this geometrical aspect contribute to the energy level splitting. First, the electrostatic potential due to the reduced positive charge of the second neighbor Ca$^{2+}$ along the $c$ axis will raise the energies of those orbitals extending along the $c$ axis, like the $yz$ and $zx$ (or $3z^2 - r^2$) states. This can be seen in Fig.3 from the global upward shift of $yz$, $zx$ bands and their oxygen bonding counterparts relative to those of the $xy$ state. Second, due to the absence of Ru-O bond at one side of the apical oxygen along the $c$ axis, the remaining Ru-O bonds are strengthened. This will further push up $yz$ and $zx$ states, which are the anti-bonding parts of the $2p$-$4d$ hybridization.

2) The compressive JT distortion. The $xy$ state is further lowered by about 0.06eV (see inset of Fig.3) due to the 2% shrinkage of the apical Ru-O bond length observed for Ca$_2$RuO$_4$ at 10K.

3) The orbital-dependent hybridization. In the AF state, the inter-site hybridization between the occupied orbitals and the unoccupied ones, which is the origin of the super-exchange, will push up the unoccupied $yz/zx$ states, and again enhance the splitting between $xy$ and $yz/zx$ in minority spin by about 0.07eV from our calculations. This effect does not exist in the FM state, and will explain the reduced occupation of $xy$ orbital in the FM solution even for $x=0.0$ (see Fig. 1(b)).

As the results, $n_{xy}^{\downarrow}$ reads 0.79, 0.72, and 0.67 for the cases corresponding to ($x=0.0$, 11K, AF), ($x=0.1$, 10K, AF: JT distortion is nearly vanishing), and ($x=0.0$, 11K, FM), respectively. It is clear that the $xy$ occupation is dominantly determined by the energy level splitting due to the 2D structure, which is comparable with the typical band width of $t_{2g}$ states. The existence of such 2D crystal-field is common for all the layered perovskites, while it has not been considered seriously so far. This effect, which is not taken into account in the analysis by Hotta and Dagotto, will certainly suppress the stability of their proposed AFO state for Ca$_2$RuO$_4$.

We also performed self-consistent calculations by including the SOC using the relativistic fully separable pseudopotentials in the framework of non-collinear magnetism. We found
that the obtained orbital occupations are almost identical to those for the case without SOC, in the AF ground state of Ca$_2$RuO$_4$. This is consistent with above discussions in the sense that the crystal field is strong enough to quench the orbital moment and stabilize the real-orbital $xy$. Note that the photoemission measurement$^4$ was done at 150K which is above the Néel temperature where the degree of orbital polarization is reduced.

The electronic structures obtained in the present work can account well also for the observed XAS$^4$ for Ca$_2$RuO$_4$ though the number of holes in the $xy$ state is not exactly 0.5 (the estimated number of holes is between 0.2 and 0.3 from our calculations for the AF ground state). There are two important aspects in the experimental analysis. One is the distribution of holes among $t_{2g}$ states and the other is its variation from the low-temperature AF phase to the high-temperature paramagnetic (PM) phase. Such information was derived from the observed relative intensity of two XAS peaks (A and B) as a function of light incidence angle $\theta$. The peaks A and B correspond to the $1s$-$2p$ transition at the apical and the in-plane oxygen, respectively. In the single electron transition limit, we obtain the XAS spectra from the converged Kohn-Sham eigen states by calculating the matrix elements of the optical transition$^{18}$. For the low-temperature AF phase, the calculation is straightforward. However, we have to introduce an approximate treatment for simulating the XAS in the high-temperature PM state. Here we use the following simplest approximation in the cluster expansion technique for treating the alloy problem$^{19}$, assuming that a magnetic state can be regarded as an alloy with two constituents, up spin state and down spin state. As the simplest approximation, we use a dimer for the cluster. Then the XAS of paramagnetic state can be approximated simply as an average of XAS of FM and AF states. (If we use a larger cluster, other magnetic orders have to be taken into account and the short-range order effect can be included.) As shown in Figs.4(a) and (c), our calculated XAS at 11K and 295K can be well compared with the experimental spectra taken at 90K and 300K (Fig.3 of Ref.$^4$). At elevated temperature, which tends to suppress the compressive JT distortion and enhance the spin disordering (increasing FM component in spin configurations), the $n_{xy}^+$ will decrease as obtained in our calculations for the 295K structure (see Figs.1(c) and (d)), being consistent with the experimental tendency.

Recently, the anisotropic optical conductivities in Ca$_2$RuO$_4$ were measured by Lee et al.$^5$ and Jung et al.$^{20}$. For the E//a spectra (see Fig.5), two peak structures (called $\alpha$ and $\beta$ at about 1.0 eV and 2.0 eV respectively) are related to the $4d - 4d$ transitions (the strong
peak at 3.0eV is due to the $p-d$ charge transfer). By increasing the temperature, spectral weight transfer from the strong peak $\beta$ to the weak peak $\alpha$ is observed and simultaneously the gap is reduced. We calculate\textsuperscript{16} the optical conductivities by using the Kubo formula\textsuperscript{21}, and all transition matrix elements are calculated from first-principles\textsuperscript{16}. As shown in Fig.5, the experimental results can be well explained by our calculations, and the corresponding transition paths are indicated in Fig.1 by red arrows. Again we use the simple average of FM and AF solutions to simulate the PM state at 295K. For the low temperature AF state, the peak $\beta$ at 2.0 eV is mostly due to the transition from the occupied majority spin $yz$ ($zx$) state at one Ru$^{4+}$ site to the unoccupied minority spin $yz$ ($zx$) state at the neighboring Ru$^{4+}$ site. In the simple energy level picture (i.e., the occupation number of each orbital is regarded as integers, either 0 or 1), it is easy to understand the origin of peak $\beta$ (which corresponds to transition shown in Fig.1(c) of Ref.\textsuperscript{5} with energy $U+J$), but not that of $\alpha$. This argument has been used by Lee et al.\textsuperscript{5} against the FO ordering. So they pursue the proposal for the AFO state\textsuperscript{12}, which has been shown in the above discussions not to be favored. However, our calculation clearly shows the existence of peak $\alpha$ at about 1.0eV, which is due to the non-vanishing transition matrix element between $xy$ and $yz/zx$ states, and crucially depends on the orthorhombic distortion and the admixture of $xy$ and $yz/zx$ states of Ru-4d. One of the strong contributions to $\alpha$ is the transition from the occupied majority spin $xy$ state at one Ru$^{4+}$ site to the unoccupied minority spin $xy$ component mixed into the $yz$, $zx$ states at the neighboring Ru$^{4+}$ site (see Fig.1(a)). Therefore, the double peak structure is just the natural consequence of strong hybridization. The peak $\beta$ should be strong at the low-temperature AF phase, and sensitive to the magnetic ordering. Actually, this transition should be suppressed in the FM configuration (as shown in Fig.1(d)), because of the flipping of the spin. The peak $\alpha$ is weak at low temperature AF state, and should be sensitive to the occupation number of $xy$ orbital. The less $n_{xy}$ is, the stronger the transition is. Actually, this transition is much enhanced in the FM configuration as shown in Fig.1(d). Therefore, with the increasing temperature, the enhancement of peak $\alpha$ can be understood by the increasing apical Ru-O bond length and the increasing FM components in the spin configuration, both of which will suppress the $xy$ occupation, while the reduction of peak $\beta$ is mostly due to the spin disordering. The reduction of the gap at elevated temperature can be understood in terms of reduction in the AF long-range order.
B. Ca$_{2-x}$Sr$_x$RuO$_4$

Above results present firm evidence for the $xy$ FO ordering for $x=0.0$. Now going from $x=0.0$ to 0.1, we see quantitative change of the electronic states (Fig.1(e) and Fig.2), while the ordering pattern does not change qualitatively. The elongation of the Ru-O bond along the $c$-axis\(^3\) (going from $x=0.0$ to 0.1) tends to suppress the occupation of $xy$ orbital, and to destroy the AF order. As a result, we see from Fig.2 the followings: 1) the energy gain of AF state with respect to the FM state reduces, being consistent with the reduced Néel temperature $T_N$ observed experimentally; 2) the reduction of ordered moments; 3) electron transfer in the minority spin channel from the $xy$ orbital ($n^{\downarrow}_{xy}$) to the $yz/zx$ orbitals ($n^{\downarrow}_{yz/zx}$); 4) redistribution of magnetization from the $yz/zx$ orbitals ($M_{yz/zx}$) to the $xy$ orbital ($M_{xy}$).

The reduction of $n^{\downarrow}_{xy}$ is also visible from the calculated XAS spectra (shown in Fig.4(b)), where the relative intensity of two peaks (A and B) is just between those of Figs.4(a) and (c).

However for $x=0.5$, where RuO$_6$ is quite elongated, and has only strong rotation around the $c$-axis without tilting, the situation is completely different (Fig.1(f) and Fig.2). These structural modifications produce basically two important changes in the electronic structure compared with the case of Ca$_2$RuO$_4$: 1) reduction in $n^{\downarrow}_{xy}$ due to the elongation; 2) broadening of the $yz/zx$ bands due to the absence of tilting. Nevertheless, the existence of RuO$_6$ rotation keeps the $xy$ band narrow\(^2\). This will contribute to the high density of states at the Fermi level of non-magnetic solution, leading to the Stoner-type FM instability. Since this instability mostly comes from the $xy$ state, we observe from the FM solution (Fig.1(f)) that the $xy$ states are strongly spin polarized, opening a pseudo-gap, and contributing to the magnetization dominantly (Fig.2). On the other hand, $yz/zx$ bands are quite broad, and are located around the Fermi level with much reduced spin polarization compared with the case of $x=0.0$. In contrast to the $S = 1$ picture with the magnetic moment supported by the $yz/zx$ orbitals for the $x=0$ case (Ca$_2$RuO$_4$), the present case may correspond to the $S=1/2$ picture where the $xy$ orbital contributes to the magnetic moment. This picture accounts well for the observations by the Curie-Weiss fitting with $S=1/2$ of susceptibility\(^2\) and the polarized neutron\(^8\) showing the spatial magnetic moment distribution of the $xy$ character. The large spin polarization for the in-plane oxygen atoms and the negligibly small one for the apical oxygen atoms (Fig.2(c)) are the results coming from the $xy$ orbital origin of the
Ru magnetic moment and are consistent with the experiment.

In contrast to the present study, the proposal in Ref.\textsuperscript{10}, which claims the contribution of $yz/zx$ states to the magnetization, was obtained by neglecting the RuO$_6$ rotation, which is an important ingredient for obtaining our orbital-dependent picture. In their treatment\textsuperscript{10}, an elaborate LDA+DMFT (dynamical mean field theory) scheme was used, but high symmetry structure of Sr$_2$RuO$_4$ was assumed. Then the effects of doping are simulated by increasing the size of $U$, instead of changing the band-width. However, we pointed out in our calculation that the strong RuO$_6$ rotation will reduce the band width of $xy$ significantly, but not that of $yz, zx$ bands. This difference produces the main source of the discrepancy discussed above.

The characteristic results of our picture for $x=0.5$ are the much suppressed (enhanced) peak A (B) in the XPS spectra (Fig.4(d)), and the structureless optical spectra below the charge-transfer peak (Fig.5(b)). These predictions should be confirmed by further experiments.

Our picture for $x=0.5$ was obtained by the LSDA calculations which tends to overestimate the FM stability. Experimentally no FM long range order has been observed, while the significant enhancement of susceptibility down to 2K clearly suggested the existence of strong FM correlation (at least for short range). Actually, the recent experiments down to 0.3K suggested the existence of weak FM component (cluster glass) at this doping. This may suggest that some magnetic orders may be energetically in near degeneracy with FM state. At present we do not have any clear idea about what magnetic orders they may be and the problem is left for future studies.

Finally, we discuss only briefly the case for the critical point $x=0.2$. 10K is already above the metal-insulator transition temperature\textsuperscript{3} in this case, and the system crystallizes in the $L-Pbca$ phase with a long apical Ru-O bond, suggesting the adjacency to the metallic side ($x=0.5$). However, in contrast to the case of $x=0.5$, the tilting of RuO$_6$ still exists to reduce the width of $yz/zx$ bands as well at $x=0.2$. This aspect tends to stabilize the insulating AF state. If we adopt LSDA, the former aspect is emphasized to make the case of $x=0.2$ very similar to the case of $x=0.5$ as can be seen in Fig.2. On the other hand, the LDA+$U$ method with $U_{\text{eff}}=2.5$ eV emphasizes the latter aspect to bring the AF state very close to the FM state with the energy difference given by $E_{\text{FM}}-E_{\text{AF}}=-3$ meV. Note, however, that the similar LDA+$U$ calculation for $x=0.5$ enhances the stability of the FM state with respect to the AF state and doubles the energy difference in Fig.2(d). These calculations, though not conclusive, clearly suggest that the system with $x=0.2$ may be in the critical
situation.

IV. CONCLUDING REMARKS

We have presented a systematic picture for the orbital states in Ca$_{2-x}$Sr$_x$RuO$_4$ ($0 \leq x \leq 0.5$). For the AF side ($x < 0.2$), we conclude that the orbital ordering is of the FO type with the dominant $xy$ occupation and that the $yz/zx$ orbitals contribute to the magnetic moment. The electronic structure corresponds basically to $S=1$ in the localized spin picture. However, strong intersite hybridization due to extended Ru 4$d$ orbitals significantly modifies the magnetic moment distribution and the present calculation agrees well with the experiments. For $x=0.5$, our LSDA calculation predicts the system to be ferromagnetic and the narrow $xy$ band with half filling may correspond to an $S=1/2$ localized spin picture. Experimentally the system shows strong tendency toward ferromagnetism but remains paramagnetic for $x=0.5$ even down to very low temperature. Only below 0.3K, the existence of cluster glass phase with weak FM component was observed. The LSDA calculation seems to overestimate the stability of ferromagnetism in other cases also. This problem is left for future studies. However, the present result for the character of the spin-density distribution is consistent with the neutron data. We emphasized that the structural modifications such as rotation, tilting and flattening of RuO$_6$ octahedron have to be fully taken into account in order to describe properly the systematic variation of the orbital states. Our results can explain other existing experiments, for example, XAS and optical conductivity, quite consistently. Furthermore, we made predictions for these experiments for the $x=0.5$ compound, which await experimental tests.

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The $xy$ band is still broad in the sense that its PDOS has a long tail in the lower energy range. However, the peak structure of PDOS around the Fermi level for the non-magnetic state is very sharp with the RuO$_6$ rotation as clearly shown in Fig.2(b) of Ref.5.
FIG. 1: The calculated PDOS of Ru-4d-t\textsubscript{2g} orbitals for various states. The solid red arrows indicate the main optical transition paths, while the dashed red arrow shows the transition which is significantly suppressed. Note the transitions indicated here mean that from the states at one Ru\textsuperscript{4+} site to the corresponding states at nearest-neighboring Ru\textsuperscript{4+} site.
FIG. 2: The calculated (a) orbital occupation numbers and (b) magnetic moments for the ground state of Ca$_{2-x}$Sr$_x$RuO$_4$ with different doping $x$. $M_{xy}$ ($M_{yz/zx}$) denotes the magnetic moment associated with the $xy$ ($yz/zx$) orbital and $M_{Ru, total} = M_{xy} + 2.0M_{yz/zx}$. The absolute value of magnetic moments from oxygens are shown in (c) (see text for further explanations). The panel (d) shows the total energy difference between the FM and AF states. The AF side ($x=0.0, 0.1$) is connected with the FM side ($x=0.2, 0.5$) by dashed lines.
FIG. 3: The projected density of states of tetragonal Ca$_2$RuO$_4$ without the JT distortion in the NM state. The inset shows the calculated energy level splitting between the $xy$ and the $yz, zx$ states for CaRuO$_3$ and Ca$_2$RuO$_4$ with tetragonal Jahn-Teller distortion, which is defined as the bond length ratios (Ru-O bond length along the $c$-axis versus that in the $ab$-plane).
FIG. 4: The calculated relative intensity of two XAS peaks (A and B located at 528.5 and 529.5 eV experimentally) as a function of light incidence angle $\theta$. 
FIG. 5: The (a) experimental\textsuperscript{20} and (b) calculated optical conductivities. The interband transitions are calculated by using the Kubo formula. The simple average of FM and AF solutions is used to simulate the PM state at 295K.