Impact of spin-orbit coupling on the magnetism of Sr$_3$MIrO$_6$ (M = Ni, Co)

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Iridates are of current great interest for their entangled spin-orbital state and possibly exotic properties. In this work, using density functional calculations, we have demonstrated that the hexagonal spin-chain materials Sr$_3$MIrO$_6$ (M = Ni, Co) are an iridate system in which the spin-orbit coupling (SOC) tunes the magnetic and electronic properties. The significant SOC alters the orbital state, the exchange pathway, and thus the magnetic structure. This work clarifies the nature and the origin of the intra-chain antiferromagnetism of Sr$_3$MIrO$_6$ and well accounts for the most recent experiments.

Charge, spin and orbital states are often coupled in 3d transition-metal oxides due to their multiple degrees of freedom and electron correlation. These states are closely related to diverse material properties and functionalities, e.g., charge ordering, orbital ordering, spin-state and magnetic transition, metal-insulator transition, superconductivity, colossal magnetoresistance, and multiferroicity. It is therefore very important to study those charge-spin-orbital states and their fascinating coupling for modeling and understanding of the abundant properties. This has formed a research stream in condensed matter physics over past decades, see e.g., a short review by Dagotto. Very recently, research interest has been extended to 5d transition-metal oxides, which probably possess a significant spin-orbit coupling and provide an avenue to novel magnetic and electronic properties due to an entangled spin-orbital state.

In this respect, iridates are a representative example. An octahedrally coordinated iridium ion normally has a large $t_{2g}$-$e_g$ crystal-field splitting due to the delocalized character of its 5d electrons. The resultant low-spin state with only a $t_{2g}$ occupation makes an open shell Ir$^{n+}$ ion (e.g., $t_{2g}^n$ for $n = 4$) behave effectively like p electrons (with an effective orbital momentum $I = 1$). As a result, an intrinsic strong spin-orbit coupling (SOC) splits the $t_{2g}$ levels into a lower $j = 3/2$ quartet and a higher $j = 1/2$ doublet. Then, for an Ir$^{n+}$ constituent oxide, the half-filled $j = 1/2$ doublet may form, due to a moderate electron correlation, a novel $j = 1/2$ Mott insulating state. It has been proposed that such a spin-orbital entangled state can bring about exotic properties, e.g., correlated topological insulator, superconductivity, Kitaev model, Weyl semimetal, and unusual magnetism.

In this Report, we have studied the 3d–5d transition-metal hybrid material Sr$_3$MIrO$_6$ (M = Ni, Co), and find that the SOC has a significant impact on its magnetism by tuning its spin-orbital states and the Ir–M inter-orbital interactions. This system has a general chemical formula A$_3$MM'O$_6$ (A = Ca, Sr; M = 3d transition metal, M' = 3d, 4d, 5d transition metal) and displays an in-plane hexagonal structure and out-of-plane spin chains, see Fig. 1. Those quasi one-dimensional spin chains each consist of alternating face-sharing MO$_6$ trigonal prisms and M'O$_6$ octahedra. This system drew a lot of attention in the past decade, because of its intriguing step-wise magnetic transition, significant Ising-like magnetism, thermoelectricity, and multiferroicity. Sr$_3$NiIrO$_6$ and Sr$_3$CoIrO$_6$ also possess fascinating magnetism. Owing to their complex temperature-dependent magnetic transitions, a standing issue is the nature of their dominant intrachain magnetism: either an intrachain ferromagnetic (FM) exchange or an antiferromagnetic (AF) coupling was proposed in previous studies. Moreover, the origin of the magnetism remains elusive. Therefore, Sr$_3$NiIrO$_6$ and Sr$_3$CoIrO$_6$ call for a prompt study to clarify the nature and origin of their intriguing intrachain magnetism. As seen below, we make a comparative study for Sr$_3$NiIrO$_6$ and Sr$_3$CoIrO$_6$ by carrying out a systematic set of electronic structure calculations. Our results consistently explain the experimental observations and settle the standing issue. In particular, we find that the SOC of the Ir$^{4+}$ ion plays an essential role in determining the intrachain AF structure of Sr$_3$MIrO$_6$ (M = Ni, Co) by tuning the crystal-field level sequence and altering the Ir–M inter-orbital interactions. Therefore, Sr$_3$MIrO$_6$ is added to the iridate category which highlights the significance of the SOC.
Results

Crystal-field levels. We first carry out spin-restricted LDA calculations to estimate the crystal field splitting. The calculated crystal-field levels can be written as $3e_g^{1.46}a_{1g}^{0.54}$, and the $a_{1g}$ level ordering can be understood, as the elongation of the IrO$_6$ octahedron along the $[111]$ direction (the resultant O-Ir-O bond angle deviating from the ideal 90° by 5.4°) raises $a_{1g}$ with respect to $e_g$ and the $e_g-e_g$ mixing pushes $e_g$ downwards.

Spin polarization and electron correlation. Then we perform spin-polarized LSDA calculations. We start with a FM or an AF state with the Ni$^{2+}$ spin $=1$ and Ir$^{4+}$ spin $=1/2$, but both calculations converge to a same FM metallic solution (not shown here). It has a total spin moment of 2.82 $\mu_B$/fu, consisting of the local spin moments of 1.46 $\mu_B$/Ni$^{2+}$, 0.54 $\mu_B$/Ir$^{4+}$, and 0.11 $\mu_B$/O. The Ni$^{2+}$ has the electronic configuration $3d^8$, and the Ir$^{4+}$ has a single $t_{2g}$ hole mostly on the $a_{1g}$ orbital [$i.e., (e_g)^3a_{1g}$]. The Ir-O and Ni-O covalencies bring about an appreciable spin moment of 0.11 $\mu_B$ on each oxygen. As the $a_{1g}$ orbital is a higher crystal-field level than the $e_g$, the $a_{1g}$ hole state allows a direct $a_{1g}$ electron hopping from Ni$^{2+}$ to Ir$^{4+}$, and this prompts the FM metallic solution.

Figure 2 | Partial density of states (DOS) of Sr$_3$NiIrO$_6$ in the nonmagnetic state calculated by LDA. The octahedral Ir ion has a common large $t_{2g} - e_g$ crystal field splitting of more than 3 eV; and in a trigonal crystal field (elongation of the IrO$_6$ octahedron along the local [111] direction, i.e., the $z$-axis of the hexagonal lattice), the $e_g$ splits further into a lower $e'_g$ doublet and a higher $a_{1g}$ singlet. The trigonal prismatic coordination produces the Ni 3d crystal-field level sequence (from low to high) as $3z^2 - r^2$, $x^2 - y^2$, and $3z^2 - r^2$. Figure 3(a) shows the spin-polarized band structure of Sr$_3$NiIrO$_6$. The calculated spin moment of Ni$^{2+}$ is 1.68 $\mu_B$, as seen below.
empty bands are too high (see Fig. 2) to be relevant for the magnetic coupling. Therefore, with the crystal-field level diagrams depicted in Figs. 4(a) and 4(b), the electron correlation and inter-orbital hybridization give rise to the FM insulating solution.

**Spin-orbit coupling.** As 5d transition metals have an intrinsic strong SOC and particularly iridates are a representative example in this respect, now we are motivated to study the SOC effect by doing LSDA + U + SOC calculations. It is interesting to note that now we get an AF insulating solution with a small band gap of 0.15 eV, see Fig. 3(b).

![Figure 3](image)

Figure 3 | The partial DOS of the Ir-5da1g and e'g orbitals in Sr3NiIrO6 in (a) the FM state calculated by LSDA + U and in (b) the AF state by LSDA + U + SOC. Taking the Ni2+ S = 1 as a reference, the Ir4+ ion has a down-spin a1g empty state in (a) but an up-spin e'g empty state (i.e., a complex orbital with \( l_z = 1 \)) in (b).

[Energy (eV)]

-2.5
-2
-1.5
-1.0
-0.5
0.0
0.5
1.0
Energy (eV)

0
1
2
3
4
5
Density of States (states/eV)

(a) FM LSDA+U

Ir 5d spin-up

Ir 5d spin-down

(b) AF LSDA+U+SOC

Ir 5d spin-up

Ir 5d spin-down

Then in the up-spin channel, the SOC lowers \( l_z = -1 \) state, and the \( l_z = 1 \) state is pushed above the Fermi level by SOC and moderate electron correlation, determining the modest band gap and giving an orbital moment of \(-0.51 \mu_B\) on Ir4+. Owing to the significant Ir-O covalency, both the spin and orbital moments are reduced from their respective ideal unit value. In the AF state of Ni2+ S = 1 and Ir4+ S = \(-1/2\), the induced magnetic moment on oxygen gets tiny (only about 0.01 \( \mu_B \)).

**Discussion**

The above results indicate an interesting evolution of the intrachain magnetic structure, from the LSDA + U FM state to the LSDA + U + SOC AF state. It is ascribed to the SOC tuning orbital state of the Ir4+ ion. Although the a1g is a higher crystal-field level than the e'g by 0.21 eV, the significant SOC of the Ir4+ ion (being about 0.5 eV) can well split the e'g doublet and eventually place the upper branch above the a1g (Fig. 4(c)). As a result, the single t2g hole of the Ir4+ ion lies in the e'g state. Then, the up-spin xz/yz electrons of the Ni2+ ion can hop, forth and back, to the up-spin e'g empty state (i.e., the up-spin \( l_z = 1 \) branch), giving rise to the AF coupling via the ddr hybridization (Figs. 4(b) and 4(c)). Actually, using the orbital states depicted in Figs. 4(b) and 4(c), we also calculated the FM state. Our LSDA + U + SOC calculations find that the FM state is indeed less stable than the AF ground state by 67 meV/f.u. Corresponding LSDA + U + SOC calculations, with Hund exchange parameter \( J = 0.9 \) eV for Ni 3d and \( J = 0.4 \) eV for Ir 5d, find that the AF ground state is more stable than the FM state by 63 meV/f.u. In addition, the spin (orbital) magnetic moment changes within 0.1 (0.05) \( \mu_B \). Therefore, here the \( J \) parameter has an insignificant influence on the calculated results. As the SOC is intrinsic in iridates, the AF ground state is deemed reliable from the LSDA + U + SOC calculations, but the FM state seems fictitious from the LSDA + U + SOC calculations without inclusion of the SOC. Indeed, the AF ground state agrees with the most recent experiment30. Therefore, we can conclude that it is the significant SOC of the Ir4+ ion which tunes the spin-orbital states and Ir-Ni inter-orbital interactions and hence determines the AF structure of Sr3NiIrO6.

Now we turn to Sr3CoIrO6. As this material has practically the same crystal structure as Sr3NiIrO6, both systems have many common features in the electronic and magnetic structures. The a1g singlet of the Ir4+ ion is higher than the e'g doublet in the crystal-field level diagram. Moreover, the high-spin Co3+ ion has the same crystal-field level sequence as Ni2+, but it now has one hole on the xz/yz doublet (compared with Ni2+, see Fig. 4(b)). Our LSDA + U calculations give a FM metallic solution due to the 3z2−r2 electron hopping and the 3/4 filled xz/yz band. Apparently, this solution contradicts the experimental AF insulating behavior30.

However, when we include SOC by doing LSDA + U + SOC calculations, we have obtained the correct AF insulating solution (see Fig. 5) in good agreement with the experiment30. The high-spin Co2+ ion (S = 3/2) has a spin moment of 2.66 \( \mu_B \). In its down-spin channel, the xz/yz−y2 doublet form the complex orbitals (x2−y2)± with \( l_z = \pm 2 \), and the Co2+ SOC lowers the \( l_z = 2 \) state but lifts the \( l_z = -2 \) state. The electron correlation makes the former at −1.5 eV and the latter at 2 eV. As a result, the Co2+ ion has also a huge orbital moment of 1.71 \( \mu_B \). In total, the Co3+ ion has the magnetic moment of 4.37 \( \mu_B \) and it is firmly aligned, due to the SOC, along the hexagonal c-axis (i.e., a significant Ising-like spin system). Moreover, the Ir 5d states are almost the same as in Sr3NiIrO6 the Ir4+ SOC places the single t2g hole on the e'g doublet; the SOC and moderate electron correlation determine the small insulating gap within the Ir4+ t2g shell, see Fig. 5. The Ir4+ ion has the spin (orbital) moment of \(-0.39 (-0.47) \mu_B\) and in total \(-0.86 \mu_B\). Using these spin-orbital states, our LSDA + U + SOC calculations find that the AF ground state is more stable than the FM state by 122 meV/f.u, which changes...
little to 130 meV/fu when including Hund exchange $J = 0.9$ eV for Co 3d and $J = 0.4$ eV for Ir 5d.

By looking at Figs. 4(b) and 4(c), and now having also one hole on the down-spin $xy$/$x^2−y^2$ doublet for Co$^{2+}$, we find that the net spin $S = 1$ from $xz/yz$ and the net spin $S = 1/2$ from $xy/x^2−y^2$ both contribute to the AF exchange with the net Ir spin $S = 1−1/2$ from the $e_g$. The former is via a $d$-d hybridization as in Sr$_2$NiIrO$_6$ and the latter a weaker $d$-d one (which is missing in Sr$_2$NiIrO$_6$). This, together with the spin values (Co$^{2+}$ $S = 3/2$ vs Ni$^{2+}$ $S = 1$), qualitatively accounts for a higher stability of the AF ground state over the FM state in Sr$_3$CoIrO$_6$ than in Sr$_3$NiIrO$_6$, i.e., 122 vs 67 meV/fu. In addition, the $d$-d hybridization results in the smaller spin and orbital moments of the Ir$^{4+}$ ion in Sr$_3$CoIrO$_6$ (0.39 + 0.47 $\mu_B$) than in Sr$_3$NiIrO$_6$ (0.44 + 0.51 $\mu_B$). Note that all these results qualitatively explain the (slightly) higher intrachain AF transition temperature of 90 K in Sr$_3$CoIrO$_6$ than 85 K in Sr$_3$NiIrO$_6$. Moreover, the calculated magnetic moments of the Ising-type, 4.37 $\mu_B$/Co$^{2+}$ and $−0.86 \mu_B$/Ir$^{4+}$, also agree reasonably well with the experimental ones of 3.6 and $−0.6 \mu_B$ in Sr$_3$CoIrO$_6$.

In summary, using density functional calculations including spin-orbit coupling (SOC) and electron correlation, we have demonstrated that the SOC of the Ir$^{4+}$ ion plays an essential role in determining the antiferromagnetism of the hexagonal spin-chain system Sr$_3$MnIrO$_6$ (M = Ni, Co) by tuning the crystal-field level sequence and altering the Ir-M inter-orbital interactions. The SOC splits the $e_g$ doublet of the octahedral Ir$^{4+}$ ion ($t_{2g}$) in a trigonal crystal field, and the single $t_{2g}$ hole resides on the $e_g$ upper branch and gives rise to the antiferromagnetic superexchange. In absence of the SOC, however, the single $t_{2g}$ hole would occupy the $t_{2g}$ singlet instead, which would mediate an unreal ferromagnetic exchange due to a direct $t_{2g}$ hopping along the Ir-M chain. We also find that the Ni$^{2+}$ and Co$^{2+}$ ions are both in a high-spin state and moreover the Co$^{2+}$ ion carries a huge orbital moment. This work well accounts for the most recent experiments and magnifies again the significance of the SOC in iridates.

**Methods**

We have carried out density functional calculations, using the full-potential augmented plane wave plus local orbital code (Wien2k 2013). We describe the Sr$_3$NiIrO$_6$ measured by neutron diffraction at 10 K and of Sr$_3$CoIrO$_6$ at 4 K. They have practically the same crystal structure: the Ir-O bond length of 2.01 Å, the M-O 2.18 Å, and the M-Ir 2.78 Å for M = Ni and Co; the small deviation of the O-Ir-O bond angle (from the ideal 90°) due to a small elongated trigonal distortion of the IrO$_6$ octahedron, being 5.4° for M = Ni and 5.2° for M = Co. The muffin-tin sphere radii are chosen to be 2.8, 2.2, 2.2 and 1.5 Bohr for Sr, Ni/Co, and Ir and O atoms, respectively. The plane-wave cut-off energy of 16 Ry is set for the interstitial wave functions, and 5 × 5 × 5 $\Gamma$ mesh for integration over the rhombohedral Brillouin zone. Using 7 × 7 × 7 $\Gamma$ mesh (more than a doubling) gives practically the same results, with the total energy converging within 2 meV/fu. We employ the local spin density approximation plus Hubbard $U$ (LSDA + U) method to describe the electron correlation of the M 3d and Ir 5d electrons, with the LSDA + U double counting correction made in a fully atomic limit. The typical values, effective $U = 2$, 4 and 5 eV ($U_{dd} = U$ with Hund exchange parameter $J$ being set at zero) are used for the Ir 5d, Co 3d, and Ni 3d states, respectively. Note that our key results—the coupled spin-orbital state and the magnetic ground state—are independent of the tested $U$ values (1–3 eV for Ir 5d and 3–7 eV for Co/Ni 3d). To account for (near) degeneracy of the Ir 5d orbitals (and of M 3d orbitals as well), the SOC is included by the second-variational method with scalar relativistic wave functions. In order to probe diverse possible spin-orbital states and magnetic structures, we excess them in our calculations by setting their respective occupation number matrix and thus orbitally dependent potentials, and then do self-consistent calculations including a full electronic relaxation. (Otherwise, some states of the concern or even the ground state cannot be achieved.) An advantage of this procedure is such that we can reliably determine the magnetic ground state by a direct comparison of the different states.

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**Author contributions**

H.W. conceived the idea and designed the research. X.O. and H.W. performed the calculations. H.W. and X.O. prepared the manuscript.

**Additional information**

**Competing financial interests** The authors declare no competing financial interests.

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