Universal excitonic superexchange in spin-orbit-coupled Mott insulators

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We point out the universal presence of the excitonic superexchange in spin-orbit-coupled Mott insulators. It is observed that, the restriction to the lowest spin-orbit-entangled “J” states may sometimes be insufficient to characterize the microscopic physics, and the virtual excitonic processes via the upper “J” states provide an important correction to the superexchange. We illustrate this excitonic superexchange from a two-dimensional 5d iridate Sr₃IrO₅ and explain its physical consequences such as the orbital-like coupling to the external magnetic flux and the nonlinear magnetic susceptibility. The universal presence of the excitonic superexchange in other spin-orbit-coupled Mott insulators such as 3d Co-based Kitaev magnets and even f electron rare-earth magnets is further discussed.

Introduction.—There has been a great interest in the field of spin-orbit-coupled correlated materials [1], ranging from the correlated spin-orbit-coupled metals or semimetals to the spin-orbit-coupled Mott insulators. The latter covers the 4d/5d rare-earth magnets like Kitaev materials, iridates, osmates [2–14], 4f rare-earth magnets [15–21], or even 3d Co-based Kitaev magnets and Ni-based diamond antiferromagnets that are of some recent interest [22–31]. In these materials, the local moments are formed by the spin-orbit-entangled “J” or “J_{eff}” states. In the study of the superexchange interactions between the local moments, the prevailing assumption was to consider the pairwise superexchange interaction between the neighboring local moments from the lowest J states, and the treatment was to carry out the perturbation theory of the extended Hubbard model. The result from this assumption and treatment were somewhat successful, especially in the systems where the lowest J states are very well separated from the excited or upper J states and at the same time the Mott gap is large.

In many of these spin-orbit-coupled Mott insulators, the spin-orbit coupling is not really a dominant energy scale especially in 3d or 4d transition metal compounds, and thus the energy separation between the lowest J states and the upper J states is not quite large compared to the hoppings and magnetic interactions, though it may involve both the crystal field effect and the spin-orbit coupling. In rare-earth magnets, this energy separation is defined by the crystal field and can sometimes be small compared to the exchange energy scale. Moreover, the famous Kitaev materials like iridates and α-RuCl₃ are not good insulator as the charge gaps in these materials are not quite large [12, 32–34]. In this work, we explore the excitonic superexchange process that involves the upper J states virtually and provide an important and universal correction to the superexchange interaction in the spin-orbit-coupled Mott insulators. The excitonic process refers to the tunneling of the electron from the lowest J states to the upper J states between the neighboring sites.

What was our previous knowledge about the superexchange interaction of the spin-orbit-coupled Mott insulators? For the J = 1/2 moments, due to the absence of the continuous rotational symmetry, all possible symmetry-allowed pairwise exchange interactions, including Heisenberg, Dzyaloshinskii-Moriya and pseudo-dipole interactions [35], appear in the exchange matrix and are likely to be equally important. Moreover, the diagonal entry of the exchange matrix from the Heisenberg and the pseudo-dipole interactions can yield the Kitaev [3] or Kitaev-like anisotropic compass interaction [2, 3]. For the larger J moments (e.g. J = 3/2 or 2), due to the larger physical Hilbert space and the strong accessibility via the spin-orbit entanglement, the pairwise interaction contains the high-order multipole interactions beyond the dipole moment interactions [4, 6, 7]. For the special quenched J moments with J = 0, the coupling with the excited J states could lead to an exciton Bose-Einstein condensation with magnetism [29, 36]. In this paper, we show that, for the unquenched local moment J, the excitonic process to the excited J states appears virtually and renders an important correction of the superexchange interaction between the local moments, especially in the presence of external magnetic field.

Atomic eigenstates.—Because of the universality and the broad applicability of this virtual excitonic process, we deliver our theory via the simplest J = 1/2 local moment for the 4d⁵ or 5d⁵ electron configuration of the Ir⁴⁺ or Ru³⁺ ion in the octahedral crystal field environment. This occurs for example in many iridates such as Sr₃IrO₅. The five degenerate atomic d₇ transitions are shown in Fig. 1(a). Only the in-plane oxygen atoms are shown. (b) The Peierls phases attached to the hopping paths, where i (i') is some nearest-neighbor (next-nearest-neighbor) hopping parameter. (c) For the spin interaction between site 1 and site 2, there are four types of hopping path for the virtual excitonic process, represented by the red triangles.

Fig. 1. (a) The square lattice of d⁷ transition metals (orange balls) with corner sharing octahedra of oxygen ligands (red balls). Only the in-plane oxygen atoms are shown. (b) The Peierls phases attached to the hopping paths, where i (i') is some nearest-neighbor (next-nearest-neighbor) hopping parameter. (c) For the spin interaction between site 1 and site 2, there are four types of hopping path for the virtual excitonic process, represented by the red triangles.
orbitals split into the $t_{2g}$ and the $e_g$ manifolds, with the former having lower energy. In the case of a large crystal field, the energy gap is large, leading to a low-spin $d^5$ configuration.

There is one hole in the $t_{2g}$ manifold which consists of orbitals $d_{yz}$, $d_{zx}$, and $d_{xy}$. The $t_{2g}$ manifold has an effective orbital angular momentum $l_{\text{eff}} = 1$ [37]. Because there is a single hole in the atomic ground state, we will use the hole representation in this paper. The atomic Hamiltonian in the $t_{2g}$ subspace at site $i$ is

$$H_0 = \sum_{m,\sigma} n^{(e)}_m \epsilon_m - \lambda \sum_{m,\sigma, \sigma'} \epsilon_{m} \mathbf{s}_{\sigma\sigma'} c^\dagger_{im\sigma} c_{im\sigma'},$$

(1)

where $c^\dagger_{im\sigma}$ ($c_{im\sigma}$) is the hole creation (annihilation) operator with orbital $m = 1, 2, 3$ and spin $\sigma = \uparrow, \downarrow$ at site $i$. The first term is the Hubbard interaction with strength $U > 0$. The prime of the summation means that the term with $(m\sigma) = (m'\sigma')$ is excluded. Note that a hole creation (annihilation) operator is an electron annihilation (creation) operator. Thus, the electron occupation number $n_{im\sigma} = 1 - n^e_{im\sigma}$ in the hole representation. The Hund’s interactions are relatively small and hence neglected. The last term is the spin-orbit interaction with strength $\lambda > 0$. In the hole representation, the sign before $\lambda$ is negative. The orbital index $m = 1, 2, 3$ corresponds to the three $t_{2g}$ orbitals $d_{yz}$, $d_{zx}$, and $d_{xy}$, respectively. In this index ordering, the orbital angular momentum matrix elements $(l_m\epsilon_n \uparrow \epsilon_{n'} \downarrow) = \delta_{mn} \epsilon_n \downarrow$, in which $l_{1,2,3}$ refers to $l_{xy,xz,yz}$, respectively. The sign is opposite to the matrix elements of the genuine $l = 1$ orbitals [37]. The spin angular momentum $s = \sigma/2$, where $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is the vector of Pauli matrices.

The eigenstates of the SOC term are two $j_{\text{eff}} = 1/2$ states with energy $-\lambda$, and four $j_{\text{eff}} = 3/2$ states with energy $\lambda/2$. In the atomic limit, the hole lies on the two-fold degenerate $j_{\text{eff}} = 1/2$ level, making the ion an effective spin-$1/2$ system. As long as the hoppings between different sites remain relatively weak as compared with the on-site Coulomb repulsion, the system remains a spin-orbit-coupled Mott insulator.

**Exchange interaction.**—The interactions between the effective spins has their origin in the virtual hopping processes. When the hoppings between different sites are taken into account, there are virtual intermediate states that contain more than one hole on the same ion site, at the cost of Coulomb repulsion energy. The intermediate states are subject to the Pauli exclusion principle, which prevents two holes with the same spin from occupying the same state. Therefore, the resulting energy correction will be dependent on the spin configurations of the initial and final states. The overall effect is the development of an effective interaction between neighboring spins, which is called exchange interaction.

In general, the hopping processes can be described by the Hamiltonian

$$T = \sum_{j,\sigma} C^\dagger_{ij\sigma} h_{ij} C_{j\sigma},$$

(2)

where $h_{ij} = h^\dagger_{ji}$ is the matrix for the hopping from site $j$ to site $i$, and $C^\dagger_{ij\sigma} = [c^\dagger_{i\sigma \uparrow}, c^\dagger_{i\sigma \downarrow}, c^\dagger_{i\sigma \uparrow}, c^\dagger_{i\sigma \downarrow} \text{,}]$ is a row vector of the $t_{2g}$ hole creation operators at site $i$ with spin $\sigma$. The Hamiltonian of the entire system is $H = H_0 + T$, where $H_0 = \sum_i H_0i$ is the summation of the atomic Hamiltonian (1) on each site. For a Mott insulator, $H_0$ dominates $T$, so the latter can be treated by perturbation theory. The matrix element of the effective Hamiltonian up to third order is

$$(H_{\text{eff}})_{mn} = (H_0)_{mn} + \sum_a T_{ma} T_{an} + \sum_{a\beta} T_{ma} T_{a\beta} T_{\beta n}$$

(3)

where the Latin indices $m$ and $n$ refer to the ground states of the unperturbed Hamiltonian $H_0$, and the Greek indices $\alpha$ and $\beta$ refer to the excited states. $\omega_{\alpha\beta}$ is the difference between the unperturbed energies of the ground state 0 and the excited state $\alpha$.

We begin by a simple model shown in Fig. 1(a), in which the $d^5$ octahedrally coordinated transition metals arrange in a two-dimensional square lattice, the same as the CuO$_2$ plane in cuprates.

By symmetry considerations, the hopping matrices for 2 $\rightarrow$ 1 and 3 $\rightarrow$ 1 in Fig. 1(a) have the form

$$h_{12} = \begin{bmatrix} t_0 & 0 & 0 \\ 0 & t_\tau & 0 \\ 0 & 0 & t_\tau \end{bmatrix} \quad \text{and} \quad h_{13} = \begin{bmatrix} \frac{\epsilon_{xy} + \epsilon_{yx}}{2} & \frac{\epsilon_{xy} - \epsilon_{yx}}{2} & 0 \\ \frac{\epsilon_{xy} - \epsilon_{yx}}{2} & \frac{\epsilon_{xy} + \epsilon_{yx}}{2} & 0 \\ 0 & 0 & \frac{3\epsilon_{xy} + \epsilon_{yx}}{4} \end{bmatrix},$$

(4)

respectively. Their subscripts hint that the hoppings are similar to the corresponding $dd$-type Slater-Koster parameters, although usually the ligand-mediated indirect hopping is the dominant mechanism. All the other hopping matrices up to second nearest neighbor can be derived by symmetry. The hopping terms for further neighbors are neglected.

Traditionally, the excited states are limited to the ones that have two holes lying on the same $j_{\text{eff}} = 1/2$ manifold, similar to the one-band Hubbard model [38]. With this restriction, only the second-order perturbation term in Eq. (3) contributes to the nearest-neighbor spin-spin interaction. By the definition of the spin operator at site $i$ as $(S_i)_{\sigma \sigma'} = c^\dagger_{i\sigma} \sigma_{\sigma'} c_{i\sigma'}$ where $s$ and $s'$ refer to the two $j_{\text{eff}} = 1/2$ states, the effective Hamiltonian for the spin interaction between site 1 and site 2 is derived as

$$H_{12} = \frac{4(2t_\tau + t_0)^2}{9U} S_1 \cdot S_2.$$ 

(5)

This is similar to the result for the one-band Hubbard model. If $t_\tau = t_0 = t$, then the coefficient $4t^2/U$ is the expected antiferromagnetic Heisenberg exchange interaction parameter.
Virtual excitonic process.—In spin-orbit-coupled Mott insulator the $j_{\text{eff}} = 1/2$ level is not well separated from the $j_{\text{eff}} = 3/2$ level, as can be seen in the typical spin-orbit-coupled Mott insulators Sr$_2$IrO$_4$ [39], Na$_2$IrO$_3$ [40], and α-RuCl$_3$ [12]. The existence of the nearby $j_{\text{eff}} = 3/2$ level will have an impact on the spin exchange interaction.

After taking the $j_{\text{eff}} = 3/2$ level into consideration, the intermediate excited states represented by the indices $\alpha$ and $\beta$ in Eq. (3) not only contain those in which two holes occupy the $j_{\text{eff}} = 1/2$ level, but also those in which one hole occupies the $j_{\text{eff}} = 1/2$ level and the other hole occupies the $j_{\text{eff}} = 3/2$ level. The process involving the latter type of excited states is called the virtual excitonic process, because it is as if one hole is excited from the $j_{\text{eff}} = 1/2$ level to the $j_{\text{eff}} = 3/2$ level, leaving an electron behind, and the virtual pair of the electron and the excited hole forms a virtual exciton.

Allowing the virtual excitonic process in the perturbation expansion (3), it is found that the correction to the two-spin exchange interaction comes from the third order term, which involves the participation of a third ion, as schematically shown in Fig. 2.

The involvement of a third ion has significant consequences when an out-of-plane magnetic field is applied. There will be additional Peierls phases in the hopping matrices. The accumulated Peierls phase for a closed loop should equal to $2\pi$ times the magnetic flux through the loop area divided by the flux quantum $\hbar c/e$. Assuming that the accumulated Peierls phase for a plaquette is $\phi$, it is convenient to use the gauge indicated in Fig. 1(b). Note that the second order interaction (5) is independent of the magnetic field because the areas of the hopping paths $1 \rightarrow 2 \rightarrow 1$ and $2 \rightarrow 1 \rightarrow 2$ are zero.

For a square lattice up to second-nearest-neighbor hopping, Fig. 1(c) shows the four hopping paths at the third perturbation order that contribute to the exchange interaction between site 1 and site 2. The resulting correction term is found to be

$$H'_{12} = \frac{16(\lambda + U)(2\delta - t_0)(\delta - t_0)(3\delta' - 2\delta - t_0)}{9U(3\lambda + 2U)^2} \cos \frac{\phi}{2} S_1 \cdot S_2 \cdot (S_2 \times S_1)$$

In sharp contrast to Eq. (5), the correction term is dependent on the magnetic field. This is because the area of the third-order hopping processes like $1 \rightarrow 2 \rightarrow 3 \rightarrow 1$ is nonzero, leading to a nonvanishing magnetic flux. Such field dependence is fundamentally different from the Zeeman interaction, because it is essentially coupled to the magnetic flux, which is an orbital effect. The above analysis can be easily generalized to different lattice models, so the effect should be ubiquitous in various spin-orbit-coupled Mott insulators.

At the third order perturbation, there are also the ring exchange interactions among each group of three neighboring spins. For the spins 1, 2, and 3 in Fig. 1(a), the ring exchange interaction is

$$H_{123} = -\frac{2(2\delta + t_0)^3}{9U^2} \sin \frac{\phi}{2} \cos \frac{\phi}{2} S_1 \cdot (S_2 \times S_3).$$

Because the mixed product of three spins breaks the time-reversal symmetry, the 3-spin ring exchange exists only when a magnetic field is applied. Hence, it must be field-dependent. The result (7) does not involve the virtual excitonic process. The hole only hops between the $j_{\text{eff}} = 1/2$ states of the three ions. The corrections by the virtual excitonic process come at the fourth perturbation order, which engages a fourth ion to provide the $j_{\text{eff}} = 3/2$ states. We expect that the correction terms have a field dependence of $\sin \phi$, because the loop area is that of a plaquette, and the product $\sin \phi S_1 \cdot (S_2 \times S_3)$ is time-reversal invariant.

Application to Sr$_2$IrO$_4$.—Having demonstrated the basic concept of the virtual excitonic process and the results on a simple model, we take a look at its effects in real material. To demonstrate it, we use Sr$_2$IrO$_4$ as an example. Sr$_2$IrO$_4$ is a well-known spin-orbit-coupled insulator [3, 38, 41]. The electron configuration of the Ir$^{4+}$ ion is $5d^5$, the same as our assumption before. It consists of corner-sharing IrO$_6$ octahedra layers with intervening Sr layers, see Fig. 3(a). The crystal structure of Sr$_2$IrO$_4$ is the Ca$_2$MnO$_4$ type with space group $I4_1/acd$ [42]. Compared to the above simple model, the complication is that the IrO$_6$ octahedra rotate alternately from perfect alignment, as shown in Fig. 3(b).

The TB model without SOC is still given by the form of Eq. (2). But we note that here the $d$ orbitals in the subscripts are defined with respect to the local coordinate system aligned with the IrO$_6$ octahedron at the corresponding site, as shown by the two sets of local coordinate systems $x'y'z'$ in Fig. 3(b). On the contrary, the spin directions are defined with respect to the global $xyz$ axes in Fig. 3(b), because in the end we would like to have a spin interaction Hamiltonian expressed in the global coordinate system.

We perform the first-principles calculations to obtain the parameters in the Hubbard Hamiltonian. Wannier functions and the tight-binding model are constructed from the Kohn-Sham orbitals, and constrained random phase approximation is used to calculate the interaction parameter $U$. See details in the Supplementary Material Supplementarymaterial.pdf

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1 The intermediate states in which two holes occupy the $j_{\text{eff}} = 3/2$ level appear only in higher order terms in the perturbative expansion.
(SM), which includes refs. [42–54].

Considering the crystal symmetry, the hole hopping matrices between the nearest-neighbor pair 12 and the second-nearest-neighbor pair 13 in Fig. 3(b) are

$$h_{12} = \begin{bmatrix} -t_3 & t_4 & 0 \\ -t_4 & -t_1 & 0 \\ 0 & 0 & -t_2 \end{bmatrix}, \quad h_{13} = \begin{bmatrix} t'_3 & -t'_4 & 0 \\ -t'_4 & t'_2 & 0 \\ 0 & 0 & -t'_1 \end{bmatrix} \tag{8}$$

respectively. The overall minus signs are to emphasize that the hopping matrices for holes differ from those for electrons by a sign, while the latter is the direct result of the first-principles calculations. We note that the zero matrix elements in $h_{12}$ and $h_{13}$ are not strict because of the interlayer interactions. However, their magnitudes are around 0.1 meV, much smaller than the other matrix elements. Thus, the smallness of these matrix elements reflects the weakness of the interlayer interactions. Other hopping matrices can be inferred by the crystal symmetry. The hopping parameters $t_{1,2,3,4}$ and $t'_{1,2,3,4}$, the SOC strength $\lambda$, and the Hubbard interaction parameter $U$ are listed in Table I.

By applying the perturbation theory, the spin interaction between the pair 12 in Fig. 3(b) is found to be

$$H_{12} = JS_1 \cdot S_2 - D(S_1 \times S_2)_z + KS_S^z S_S^z - \left[J'S_1 \cdot S_2 + D'(S_1 \times S_2)_z - K'S_S^z S_S^z \right] \cos \frac{\phi}{2}. \tag{9}$$

The analytical expressions for the coefficients are given in the SM, and their numerical values are listed in Table I. The terms involving $J'$, $D'$, and $K'$ come from the virtual excitonic processes. The phase $\phi = eB|z|/\hbar$, where $B$ is the out-of-plane magnetic field, and $l = 3.88$ Å is the distance between nearest-neighbor Ir atoms. The order of magnitude $\phi \sim 1$ corresponds to a magnetic field $B \sim 4.4 \times 10^3$ T.

Nonlinear susceptibility.—The positive coefficient before $S_S^z S_S^z$ leads to the easy-plane anisotropy. The in-plane magnetic structure is cantent antiferromagnetic because of the competition between the Heisenberg and the Dzyaloshinskii-Moriya interaction (the first two terms of Eq. (9)). With the application of a magnetic field $B$ perpendicular to the IrO$_2$ layers, the cantent antiferromagnetic structure develops an out-of-plane component. We first neglect the virtual excitonic process corrections, then the classical energy per site is

$$E = 2 \left[JS_A \cdot S_B - D(S_A \times S_B)_z + KS_A^z S_B^z - 2\mu_B BS_A^z \right] - 2 \mu_B BS_A^z, \tag{10}$$

where $S_A$ and $S_B$ are the spins on the two sublattices, with the same $z$ component, and $\mu_B$ is the Bohr magneton. We have used the fact that the Landé g-factor of the $j_{eff} = 1/2$ hole state is $-2$. By minimizing the energy, the tilting angle of the spins away from the $xy$ plane can be obtained, and the perpendicular magnetic susceptibility is found to be

$$\chi_{\perp} = \frac{N\mu_B^2}{\sqrt{J^2 + D^2} + J + K}, \tag{11}$$

in which $N$ is the site density. This result (11) has the same property as the mean-field result for the Néel antiferromagnetism that below the Néel temperature it depends neither on the temperature nor on the magnetic field.

Now we take into account the correction term from the virtual excitonic process. It amounts to the replacements $J \rightarrow J - J' \cos(\phi/2)$, $D \rightarrow D + D' \cos(\phi/2)$, and $K \rightarrow K + K' \cos(\phi/2)$. With Taylor expansion up to $O(\phi^2)$, we find the magnetization

$$M = \chi_{\perp}^{(1)} B + \chi_{\perp}^{(3)} B^3 + \cdots, \tag{12}$$

where

$$\chi_{\perp}^{(1)} = \frac{N\mu_B^2}{\sqrt{(J - J')^2 + (D + D')^2} + J - J' + K + K'}, \tag{13}$$

is linear susceptibility, and

$$\chi_{\perp}^{(3)} = -\frac{N\mu_B^2}{8 \left[\sqrt{(J - J')^2 + (D + D')^2} + J - J' + K + K'\right]^2} \left(\frac{e^2}{\hbar}\right)^2 \tag{14}$$

is the nonlinear susceptibility [55, 56]. Its existence is the consequence of the virtual excitonic process. This clarifies one important source of the nonlinear susceptibility in these systems.

Discussion.—Here we discuss the applicability of the excitonic superexchange. We have shown that, the virtual excitonic process appears in the high order perturbation theory of the multiple-band Hubbard model. A smaller spin-orbit coupling and/or a weak Hubbard interaction could enhance its contribution. Apparently, in many of these 4$d$/5$d$ magnets, the spin-orbit coupling is not really the dominant energy scale, and the systems also behave like weak Mott insulators. The 4$d$ magnet $\alpha$-RuCl$_3$ is a good example of this kind. This material shows a remarkable thermal Hall transport result in external magnetic fields, which is likely to be related to the gapped Kitaev spin liquid [57–59]. More recently, it has been found that its thermal conductivity oscillates when an external magnetic field is applied [60]. It is believed that the oscillation originates from the existence of the spinon Fermi surface of some kind. The orbital quantization of the spinons requires the coupling of the spinons with an internal U(1) gauge field [61]. Such orbital effect is obviously not controlled via the Zeeman term. The virtual excitonic process and/or the strong charge
fluctuation could provide the microscopic means to lock the internal U(1) gauge field with the external magnetic flux, and thereby generating the oscillatory behaviors.

For the Co-based Kitaev materials [22–26], the Hubbard interaction is large, but the spin-orbit coupling is weak. Thus, the excitonic superexchange may still be important. For the rare-earth magnets, the crystal field enters as an important energy scale. If the crystal field energy separation between the ground states and the excited states is not large enough compared to the exchange energy scale, the excited states would be involved into the low-temperature magnetic physics. This upper-branch magnetism has been invoked for the pyrochlore magnet Tb$_2$Ti$_2$O$_7$ [62–65].

The concept could also be applied to systems such as metal organic frameworks, molecular magnets, and Moiré systems, which have large lattice constants. Another example is the spin-liquid candidate material 1T-TaS$_2$ [66–70], which enters into a charge-density-wave (CDW) phase at low temperature. In the CDW phase, each supercell has a localized unpaired electron which provides the spin. The neighboring spins are separated at a distance $l$ larger than ten angstroms. Moreover, the orbitals $d_{xy}$, $d_{yz}$, and $d_{zx}$ have similar energies [71], which facilitates the excitonic process. The required magnetic field $B$ for the phase $\phi \sim eB^2/\hbar$ to be at the order of $O(1)$ drops to hundreds of Tesla, making the effect of excitonic process more prominent.

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[1] W. Witzczak-Krempa, G. Chen, Y. B. Kim, and L. Balents, Correlated quantum phenomena in the strong spin-orbit regime, *Annual Review of Condensed Matter Physics* **5**, 57–82 (2014).

[2] G. Chen and L. Balents, Spin-orbit effects in Na$_2$IrO$_3$: A hyper-kagome lattice antiferromagnet, *Phys. Rev. B* **78**, 094403 (2008).

[3] G. Jackeli and G. Khaliullin, Mott insulators in the strong spin-orbit coupling limit: From heisenberg to a quantum compass and kitaev models, *Phys. Rev. Lett.* **102**, 017205 (2009).

[4] G. Chen, R. Pereira, and L. Balents, Exotic phases induced by strong spin-orbit coupling in ordered double perovskites, *Phys. Rev. B* **82**, 174440 (2010).

[5] J. c. v. Chaloupka, G. Jackeli, and G. Khaliullin, Kitaev-Heisenberg Model on a Honeycomb Lattice: Possible Exotic Phases in Iridium Oxides $A_2$IrO$_3$, *Phys. Rev. Lett.* **105**, 027204 (2010).

[6] G. Chen and L. Balents, Spin-orbit coupling in $d^3$ ordered double perovskites, *Phys. Rev. B* **84**, 094420 (2011).

[7] G. Chen and L. Balents, Erratum: Spin-orbit coupling in $d^3$ ordered double perovskites [Phys. Rev. B 84, 094420 (2011)], *Phys. Rev. B* **91**, 219903 (2015).

[8] J. c. v. Chaloupka, G. Jackeli, and G. Khaliullin, Zigzag Magnetic Order in the Iridium Oxide Na$_2$IrO$_3$, *Phys. Rev. Lett.* **110**, 097204 (2013).

[9] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Generic Spin Model for the Honeycomb Iridates beyond the Kitaev Limit, *Phys. Rev. Lett.* **112**, 077204 (2014).

[10] H.-S. Kim, V. S. V., A. Catuneanu, and H.-Y. Kee, Kitaev magnetism in honeycomb RuCl$_3$ with intermediate spin-orbit coupling, *Phys. Rev. B* **91**, 241110 (2015).

[11] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Spin-orbit physics giving rise to novel phases in correlated systems: Iridates and related materials, *Annual Review of Condensed Matter Physics* **7**, 195–221 (2016).

[12] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Lee, and Y.-J. Kim, $\sigma \sim \text{RuCl}_3$: A spin-orbit assisted Mott insulator on a honeycomb lattice, *Phys. Rev. B* **90**, 041112 (2014).

[13] S. Trebst, Kitaev materials (2017), arXiv:1701.07056 [cond-mat.str-el];

[14] M. Hermanns, I. Kimchi, and J. Knolle, Physics of the kitaev model: Fractionalization, dynamic correlations, and material connections, *Annual Review of Condensed Matter Physics* **9**, 17–33 (2018).

[15] J. G. Rau and M. J. Gingras, Frustrated Quantum Rare-Earth Pyrochlores, *Annual Review of Condensed Matter Physics* **10**, 357–386 (2019).

[16] Y.-D. Li, X. Wang, and G. Chen, Anisotropic spin model of strong spin-orbit-coupled triangular antiferromagnets, *Phys. Rev. B* **94**, 035107 (2016).

[17] C. Liu, C.-J. Huang, and G. Chen, Intrinsic quantum Ising model on a triangular lattice magnet $\text{TmMgGaO}_4$, *Phys. Rev. Research* **2**, 043013 (2020).

[18] Y.-D. Li and G. Chen, Symmetry enriched $U(1)$ topological orders for dipole-octupole doublets on a pyrochlore lattice, *Phys. Rev. B* **95**, 041106 (2017).

[19] Y.-D. Li, X. Wang, and G. Chen, Hidden multipolar orders of dipole-octupole doublets on a triangular lattice, *Phys. Rev. B* **94**, 201114 (2016).

[20] L. Savary, X. Wang, H.-Y. Kee, Y. B. Kim, Y. Yu, and G. Chen, Quantum spin ice on the breathing pyrochlore lattice, *Phys. Rev. B* **94**, 075146 (2016).

[21] J. G. Rau, L. S. Wu, A. F. May, L. Poudel, B. Winn, V. O. Garlea, A. Huq, P. Whitfield, A. E. Taylor, M. D. Lumsden, M. J. P. Gingras, and A. D. Christianson, Anisotropic Exchange within Decoupled Tetrahedra in the Quantum Breathing Pyrochlore $\text{Ba}_3\text{Yb}_2\text{Zn}_2\text{O}_{11}$, *Phys. Rev. Lett.* **116**, 257204 (2016).

[22] H. Liu, J. Chaloupka, and G. Khaliullin, Kitaev Spin Liquid in 3d Transition Metal Compounds, *Phys. Rev. Lett.* **125**, 047201 (2020).

[23] S. Das, S. Voleti, T. Saha-Dasgupta, and A. Paramekanti, Xy magnetism, kitaev exchange, and long-range frustration in the $J_{eff} = \frac{1}{2}$ honeycomb cobaltates, *Phys. Rev. B* **104**, 134425 (2021).

[24] H. Liu, Towards kitaev spin liquid in 3d transition metal compounds, *International Journal of Modern Physics B* **35**, 2130006 (2021).

[25] H. Liu and G. Khaliullin, Pseudospin exchange interactions in $d^3$ cobalt compounds: Possible realization of the kitaev model, *Phys. Rev. B* **97**, 014407 (2018).
[26] R. Sano, Y. Kato, and Y. Motome, Kitaev-Heisenberg Hamiltonian for high-spin $d^7$ Mott insulators, Phys. Rev. B 97, 014408 (2018).
[27] J. R. Chamorro, L. Ge, J. Flynn, M. A. Subramanian, M. Mourigal, and T. M. McQueen, Frustrated spin one on a diamond lattice in NiRh$_2$O$_4$, Phys. Rev. Materials 2, 034404 (2018).
[28] G. Chen, Quantum paramagnet and frustrated quantum criticality in a spin-one diamond lattice antiferromagnet, Phys. Rev. B 96, 020412 (2017).
[29] F.-Y. Li and G. Chen, Spin-orbital entanglement in $d^9$ Mott insulators: Possible excitonic magnetism in diamond lattice antiferromagnets, Phys. Rev. B 100, 045103 (2019).
[30] F. L. Buesen, M. Hering, J. Reuther, and S. Trebst, Quantum spin liquids in frustrated spin-1 diamond antiferromagnets, Phys. Rev. Lett. 120, 057201 (2018).
[31] S. Das, D. Nafday, T. Saha-Dasgupta, and A. Paramekanti, NiRh$_2$O$_4$: A spin-orbit entangled diamond-lattice paramagnet, Phys. Rev. B 100, 140408 (2019).
[32] Y. H. Gao, C. Hickey, T. Xiang, S. Trebst, and G. Chen, Thermal hall signatures of non-kitaev spin liquids in honeycomb kitaev materials, Phys. Rev. Research 1, 013014 (2019).
[33] X. Zhou, H. Li, J. A. Waugh, S. Parham, H.-S. Kim, J. A. Sears, A. Gomes, H.-Y. Kee, Y.-J. Kim, and D. S. Dessau, Angle-resolved photoemission study of the Kitaev candidate $\alpha$-RuCl$_3$, Phys. Rev. B 94, 161106 (2016).
[34] L. J. Sandilands, Y. Tian, A. A. Reinders, H.-S. Kim, K. W. Plumb, Y.-J. Kim, H.-Y. Kee, and K. S. Burch, Spin-orbit excitations and electronic structure of the putative Kitaev magnet $\alpha$–RuCl$_3$, Phys. Rev. B 93, 075144 (2016).
[35] S. Maejawa, T. Tohyama, S. Barnes, S. Ishihara, W. Koshihai, and G. Khaliullin, Physics of Transition Metal Oxides (Springer-Verlag Berlin Heidelberg, 2003).
[36] G. Khaliullin, Excitonic Magnetism in Van Vleck–type $d^9$ Mott Insulators, Phys. Rev. Lett. 111, 197201 (2013).
[37] A. Abramag and B. Bleaney, Electron Paramagnetic Resonance of Transition Ions (Oxford University Press, 1970).
[38] F. Wang and T. Senthil, Twisted Hubbard Model for Sr$_2$IrO$_4$: Magnetism and Possible High Temperature Superconductivity, Phys. Rev. Lett. 106, 136402 (2011).
[39] B. J. Kim, H. Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, J. Yu, T. W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg, Novel $J_{\text{eff}} = 1/2$ Mott State Induced by Relativistic Spin-Orbit Coupling in Sr$_2$IrO$_4$, Phys. Rev. Lett. 101, 076402 (2008).
[40] R. Comin, G. Levy, B. Ludbrook, Z.-H. Zhu, C. N. Veenastra, J. A. Rosen, Y. Singh, P. Gegenwart, D. Stricker, J. N. Hancock, D. van der Marel, I. S. Elfimov, and A. Damascelli, Na$_2$IrO$_3$ as a Novel Relativistic Mott Insulator with a 340-meV Gap, Phys. Rev. Lett. 109, 266402 (2012).
[41] L. Engström, T. Peregr-Barnea, and W. Witzczak-Krempa, Modelling multiorbital effects in Sr$_2$IrO$_4$ under strain and a Zeeman field, Phys. Rev. B 103, 155147 (2021).
[42] Q. Huang, J. Soubeyroux, O. Chmaissem, I. Sora, A. Santoro, R. Cava, J. Krajewski, and W. Peck, Neutron Powder Diffraction Study of the Crystal Structures of Sr$_2$RuO$_4$ and Sr$_2$IrO$_4$ at Room Temperature and at 10 K, Journal of Solid State Chemistry 112, 355 (1994).
[43] N. Marzari and D. Vanderbilt, Maximally localized generalized wannier functions for composite energy bands, Phys. Rev. B 56, 12847 (1997).
[44] I. Souza, N. Marzari, and D. Vanderbilt, Maximally localized wannier functions for entangled energy bands, Phys. Rev. B 65, 035109 (2001).
[45] P. Hohenberg and W. Kohn, Inhomogeneous Electron Gas, Phys. Rev. 136, B864 (1964).
[46] W. Kohn and L. J. Sham, Self-Consistent Equations Including Exchange and Correlation Effects, Phys. Rev. 140, A1133 (1965).
[47] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, Quantum espresso: a modular and open-source software project for quantum simulations of materials, Journal of Physics: Condensed Matter 21, 395502 (19pp) (2009).
[48] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunka, M. B. Nardielli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni, N. Colonna, I. Carmimeo, A. D. Corso, S. de Gironcoli, P. Delugas, R. A. D. Jr, A. Ferretti, A. Floris, G. Fratesi, G. Fugallo, R. Gebauer, U. Gerstmann, F. Giustino, T. Gorni, J. Jia, M. Kawamura, H.-Y. Ko, A. Kokalj, E. K¨uct¨ubkenli, M. Lazzeri, M. Marsili, N. Marzari, F. Mauri, N. L. Nguyen, H.-V. Nguyen, A. O. de la Roa, L. Paulatto, S. Foncé, D. Rocca, R. Sabatini, B. Santra, M. Schlipf, A. P. Seitsonen, A. Smogunov, I. Timrov, T. Thonhauser, P. Umari, N. Vasi, X. Wu, and S. Baroni, Advanced capabilities for materials modelling with quantum espresso, Journal of Physics: Condensed Matter 29, 465901 (2017).
[49] M. Schlipf and F. Gygi, Optimization algorithm for the generation of ONCV pseudopotentials, Computer Physics Communications 196, 36 (2015).
[50] D. R. Hamann, Optimized norm-conserving Vanderbilt pseudopotentials, Phys. Rev. B 88, 085117 (2013).
[51] F. Aryasetiawan, T. Miyake, and R. Sakuma, The constrained RPA method for calculating the hubbard U from first-principles, in The LDA+DMFT approach to strongly correlated materials, edited by E. Pavarini, E. Koch, D. Vollhardt, and A. Lichtenstein (Forschungszentrum Jülich GmbH, Institute for Advanced Simulations, Jülich, 2011).
[52] K. Nakamura, Y. Yoshimoto, Y. Nomura, T. Tadano, M. Kawamura, T. Kosugi, K. Yoshimi, T. Misawa, and Y. Motoyama, RESPACK: An ab initio tool for derivation of effective low-energy model of material, Computer Physics Communications 261, 107781 (2021).
[53] Y. Hinuma, G. Pizzi, Y. Kumagai, F. Oba, and I. Tanaka, Band structure diagram paths based on crystallography, Computational Materials Science 128, 140 (2017).
[54] A. Togo and I. Tanaka, Spglib: a software library for crystal symmetry search (2018), arXiv:1808.01590 [cond-mat.mtrl-sci].
[55] S. Fujiki and S. Katsura, Nonlinear Susceptibility in the Spin Glass, Progress of Theoretical Physics 65, 1130 (1981), https://academic.oup.com/ptp/article-pdf/65/4/1130/5427375/65-4-1130.pdf.
[56] Y. Machida, S. Nakatsuji, S. Onoda, T. Tayama, and T. Sakakibara, Time-reversal symmetry breaking and spontaneous hall effect without magnetic dipole order, Nature 463, 210 (2010).
[57] T. Yokoi, S. Ma, Y. Kasahara, S. Kasahara, T. Shibaudchi, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, C. Hickey, and et al., Half-integral quantized anomalous thermal Hall effect in the Kitaev material candidate $\alpha$-RuCl$_3$, Science 373, 568–572 (2021).
[58] Y. Kasahara, K. Sugii, T. Ohnishi, M. Shimozawa, M. Yamashita, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibaudchi, and et al., Unusual Thermal Hall Effect in a Kitaev
Spin Liquid Candidate $\alpha$-RuCl$_3$, Phys. Rev. Lett. 120, 217205 (2018).

[59] Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, S. Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, and et al., Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid, Nature 559, 227–231 (2018).

[60] P. Czajka, T. Gao, M. Hirschberger, P. Lampen-Kelley, A. Banerjee, J. Yan, D. G. Mandrus, S. E. Nagler, and N. P. Ong, Oscillations of the thermal conductivity in the spin-liquid state of $\alpha$-RuCl$_3$, Nature Physics 17, 915 (2021).

[61] O. I. Motrunich, Orbital magnetic field effects in spin liquid with spinon Fermi sea: Possible application to $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$, Phys. Rev. B 73, 155115 (2006).

[62] B. D. Gaulin, J. S. Gardner, P. A. McClarty, and M. J. P. Gingras, Lack of evidence for a singlet crystal-field ground state in the magnetic pyrochlore Tb$_2$Ti$_2$O$_7$, Phys. Rev. B 84, 140402 (2011).

[63] H. R. Molavian, M. J. P. Gingras, and B. Canals, Dynamically Induced Frustration as a Route to a Quantum Spin Ice State in Tb$_2$Ti$_2$O$_7$ via Virtual Crystal Field Excitations and Quantum Many-Body Effects, Phys. Rev. Lett. 98, 157204 (2007).

[64] C. Liu, F.-Y. Li, and G. Chen, Upper branch magnetism in quantum magnets: Collapses of excited levels and emergent selection rules, Phys. Rev. B 99, 224407 (2019).

[65] H. Kadowaki, M. Wakita, B. Fäk, J. Ollivier, and S. Ohira-Kawamura, Spin and quadrupole correlations by three-spin interaction in the frustrated pyrochlore magnet Tb$_{2+\delta}$Ti$_{2-\delta}$O$_{7+\epsilon}$, Phys. Rev. B 105, 014439 (2022).

[66] K. T. Law and P. A. Lee, 1T-TaS$_2$ as a quantum spin liquid, Proceedings of the National Academy of Sciences 114, 6996 (2017).

[67] W.-Y. He, X. Y. Xu, G. Chen, K. T. Law, and P. A. Lee, Spinon Fermi Surface in a Cluster Mott Insulator Model on a Triangular Lattice and Possible Application to 1T–TaS$_2$, Phys. Rev. Lett. 121, 046401 (2018).

[68] C. J. Butler, M. Yoshida, T. Hanaguri, and Y. Iwasa, Mottness versus unit-cell doubling as the driver of the insulating state in 1T-TaS$_2$, Nature Communications 11, 4215 (2020).

[69] Y. D. Wang, W. L. Yao, Z. M. Xin, T. T. Han, Z. G. Wang, L. Chen, C. Cai, Y. Li, and Y. Zhang, Band insulator to Mott insulator transition in 1T-TaS$_2$, Nature Communications 11, 2477 (2020).

[70] C.-K. Li, X.-P. Yao, J. Liu, and G. Chen, Fractionalization on the surface: Is type-ii terminated 1T–TaS$_2$ surface an anomalously realized spin liquid?, Phys. Rev. Lett. 129, 017202 (2022).

[71] K. Rossnagel and N. V. Smith, Spin-orbit coupling in the band structure of reconstructed 1T–TaS$_2$, Phys. Rev. B 73, 073106 (2006).
I. DETAILS OF THE FIRST-PRINCIPLES CALCULATIONS

To obtain the hopping parameters of Sr$_2$IrO$_4$, we use first-principles calculations to get the Kohn-Sham (KS) states and the eigenenergies. Then the maximally localized Wannier functions (MLWFs) [1, 2] are constructed, and the tight-binding (TB) model is established. The first-principles calculations are based on density functional theory [3, 4] and plane-wave pseudopotential method, as implemented in the QUANTUM ESPRESSO package [5, 6]. The low temperature experimental crystal structure [7] is adopted in the calculations. The energy cutoff of the plane wave basis set is 120 Ry. The Brillouin zone is sampled by a $6 \times 6 \times 6 \ \text{k}$-grid. The Schlipf-Gygi norm-conserving pseudopotentials [8] produced by the ONCVPSP code [9] are used. The MLWFs and the TB model are constructed by the RESPACK program [10]. The initial guesses of the Wannier functions (WFs) are the projections of the KS states near the Fermi level to the local $t_{2g}$ orbitals of each Ir atom. The spatial spread of the WFs are then minimized.

Fig. S1. The Brillouin zone and the high symmetry paths of Sr$_2$IrO$_4$.
The comparison of the band structures calculated by the TB model and DFT without spin-orbit coupling (SOC) is presented in Fig. S2(a). The high symmetry paths in the Brillouin zone shown in Fig. S1 are generated by the SeeK-path tool [11, 12]. It can be seen that the tight binding model reproduces the DFT results perfectly. The nearest-neighbor and next-nearest-neighbor hopping parameters are listed in Table I of the main text.

We then add to the obtained TB model a local SOC term (the second term of Eq. (1) in the main text), and compare its band structure with the DFT result including SOC. The SOC strength $\lambda$ is fitted by the least square method to be $\lambda = 0.36$ eV. The corresponding band structures are shown in Fig. S2(b). The details near the Fermi level ($E = 0$) are well reproduced by the TB model with the local SOC term.

To evaluate the Hubbard parameter $U$, constrained random phase approximation [13] is adopted, as implemented in the RESPACK program [10]. The Hilbert space is divided into the subspace of the $t_{2g}$ orbitals in which the MLWFs are constructed, and the rest subspace ($r$ subspace). $U$ is taken to be the orbital-averaged on-site static Coulomb interaction partially screened by the polarization processes involving the $r$ subspace. It is found that $U = 2.232$ eV.

II. ANALYTICAL EXPRESSIONS FOR THE SPIN INTERACTION PARAMETERS

The analytical expressions for the spin interaction parameters of Sr$_2$IrO$_4$ in the main text are

$$ J = \frac{4\left[(t_1 + t_2 + t_3)^2 - 4t_4^2\right]}{9U}, $$

(1)
TABLE S1. The parameters of the spin interaction between site 1 and site 2 in Fig. 3(b) of the main text, in meV. The nonvanishing tetragonal crystal field is taken into account.

|     |     |     |     |     |     |
|-----|-----|-----|-----|-----|-----|
| J   | D   | K   | J'  | D'  | K'  |
| 59.3| 14.2| 1.67| 2.56| 0.597| 0.209|

III. EFFECTS OF THE TETRAGONAL CRYSTAL FIELD

In deriving the above analytical expressions, we have assumed a perfect octahedral environment of the Ir\(^{4+}\) ion. As a result, the three \(t_{2g}\) orbitals are degenerate. In the crystal of Sr\(_2\)IrO\(_4\), their is a tetragonal crystal field which makes the energy of the \(d_{xy}\) orbital different from that of the degenerate \(d_{xz}\) and \(d_{yz}\) orbitals. It gives rise to an additional term in the atomic Hamiltonian

\[
H_{CEF}^{0i} = \sum_\sigma C_{i\sigma}^\dagger \begin{bmatrix} \Delta & 0 & 0 \\ 0 & \Delta & 0 \\ 0 & 0 & 0 \end{bmatrix} C_{i\sigma},
\]

where \(C_{i\sigma}^\dagger = [c_{i,yz,\sigma}^\dagger, c_{i,zx,\sigma}^\dagger, c_{i,xy,\sigma}^\dagger]\) as in the main text. It is found that \(\Delta = 159\) meV.

There are no simple analytical expressions for the spin interaction parameters for nonvanishing \(\Delta\). The numerical results are listed in Table S1. Compared with the corresponding parameters in the Table I of the main text, it can be seen that the tetragonal crystal field has only minor influences on the spin interactions.
[1] N. Marzari and D. Vanderbilt, Maximally localized generalized wannier functions for composite energy bands, Phys. Rev. B 56, 12847 (1997).

[2] I. Souza, N. Marzari, and D. Vanderbilt, Maximally localized wannier functions for entangled energy bands, Phys. Rev. B 65, 035109 (2001).

[3] P. Hohenberg and W. Kohn, Inhomogeneous Electron Gas, Phys. Rev. 136, B864 (1964).

[4] W. Kohn and L. J. Sham, Self-Consistent Equations Including Exchange and Correlation Effects, Phys. Rev. 140, A1133 (1965).

[5] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, Quantum espresso: a modular and open-source software project for quantum simulations of materials, Journal of Physics: Condensed Matter 21, 395502 (19pp) (2009).

[6] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunau, M. B. Nardelli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni, N. Colonna, I. Carnimeo, A. D. Corso, S. de Gironcoli, P. Delugas, R. A. D. Jr, A. Ferretti, A. Floris, G. Fratesi, G. Fugallo, R. Gebauer, U. Gerstmann, F. Giustino, T. Gorni, J. Jia, M. Kawamura, H.-Y. Ko, A. Kokalj, E. Küçükbenli, M. Lazzeri, M. Marsili, N. Marzari, F. Mauri, N. L. Nguyen, H.-V. Nguyen, A. O. de-la Roza, L. Paulatto, S. Poncé, D. Rocca, R. Sabatini, B. Santra, M. Schlipf, A. P. Seitsonen, A. Smogunov, I. Timrov, T. Thonhauser, P. Umari, N. Vast, X. Wu, and S. Baroni, Advanced capabilities for materials modelling with quantum espresso, Journal of Physics: Condensed Matter 29, 465901 (2017).

[7] Q. Huang, J. Soubeyroux, O. Chmaissem, I. Sora, A. Santoro, R. Cava, J. Krajewski, and W. Peck, Neutron Powder Diffraction Study of the Crystal Structures of Sr$_2$RuO$_4$ and Sr$_2$IrO$_4$ at Room Temperature and at 10 K, Journal of Solid State Chemistry 112, 355 (1994).

[8] M. Schlipf and F. Gygi, Optimization algorithm for the generation of ONCV pseudopotentials, Computer Physics Communications 196, 36 (2015).

[9] D. R. Hamann, Optimized norm-conserving Vanderbilt pseudopotentials, Phys. Rev. B 88, 085117 (2013).
[10] K. Nakamura, Y. Yoshimoto, Y. Nomura, T. Tadano, M. Kawamura, T. Kosugi, K. Yoshimi, T. Misawa, and Y. Motoyama, RESPACK: An ab initio tool for derivation of effective low-energy model of material, Computer Physics Communications 261, 107781 (2021).

[11] Y. Hinuma, G. Pizzi, Y. Kumagai, F. Oba, and I. Tanaka, Band structure diagram paths based on crystallography, Computational Materials Science 128, 140 (2017).

[12] A. Togo and I. Tanaka, Spglib: a software library for crystal symmetry search (2018), arXiv:1808.01590 [cond-mat.mtrl-sci].

[13] F. Aryasetiawan, T. Miyake, and R. Sakuma, The constrained RPA method for calculating the hubbard U from first-principles, in The LDA+DMFT approach to strongly correlated materials, edited by E. Pavarini, E. Koch, D. Vollhardt, and A. Lichtenstein (2011).