Topological edge states in single layers of honeycomb materials with strong spin-orbit coupling

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We study possible edge states in single layers of honeycomb materials such as α-RuCl3 and A2IrO3 (A=Li, Na) with strong spin-orbit coupling (SOC). These two-dimensional systems exhibit linearly dispersing one-dimensional (1D) edge states when their 1D boundary forms a zig-zag shape. Using an effective tight-binding model based on first principles band structure calculations including Hubbard U and SOC, we find degenerate edge states at the zone center and zone boundary. The roles of chiral symmetry and time-reversal symmetry are presented. The implications to experimental signatures and the effects of disorder are also discussed.

Introduction — The honeycomb lattice has provided an excellent playground to explore novel physics in both weakly and strongly correlated systems. Various phases such as the Dirac semimetal[1], Chern insulator[2], and quantum spin Hall (QSH) insulator[3, 4] have been proposed in honeycomb lattices with and without spin-orbit coupling (SOC). Moreover, strongly correlated honeycomb systems such as A2IrO3 (A = Na or Li)[5], in which the multi-orbital t2g model is mapped into a single orbital with pseudospin jeff = 1/2, could in principle realize the exactly solvable Kitaev spin model[8–12]. Along this vein of thinking, α-RuCl3 (RuCl3) was recently proposed as another promising candidate to exhibit Kitaev physics[13, 14].

The unconventional magnetic ordering reported in RuCl3, Na2IrO3, and Li2IrO3[5, 11, 15–21] can be understood as a competition between Kitaev and other exchange terms[22–24] and thus the honeycomb lattice endowed with strong SOC has been one of hot topics in the community of spin liquid and topological phases.

From the topological point of view, it was suggested that honeycomb Na2IrO3 can support the quantum spin Hall (QSH) state above the magnetic ordering temperature via complex second nearest neighbor hopping terms due to a combination of strong SOC and the edge sharing nature of the oxygen octahedra[25], which leads to a gap at the K- and K’-point[25]. This idea is an extension of the QSH in graphene[3], but it should be easier to observe in iridates because the SOC is much stronger. Interestingly, an angle-resolved photoemission spectroscopy (ARPES) measurement on Na2IrO3 at room temperature reported linearly dispersing bands, but at the Γ-point[26]. This observation remains unsolved, since a theoretical study based on ab-initio calculation found a trivial topological Z2 invariant for Na2IrO3[27], and there is currently no microscopic theory that supports such a surface state in the compound. Furthermore, other ARPES experiments[28, 29] did not find any linearly dispersing bands in Na2IrO3, perhaps indicating that differences in sample surfaces could be giving contrary results.

Here we study edge states of two-dimensional single layers of α-RuCl3 and A2IrO3 by employing ab-initio electronic structure calculations including SOC and the Hubbard interaction U of 2-3eV, and analyzing an effective tight-binding (TB) model based on jeff = 1/2 pseudospins. While the Hubbard interaction of 2-3eV favors a magnetic ground state, we impose a paramagnetic condition to study the topological nature of these materials in the absence of magnetic order, which is thus applicable to the region above the magnetic ordering temperature. Furthermore, the Hubbard U enhances the jeff = 1/2 picture and makes the band gap bigger than without U[14]. We estimate jeff = 1/2 TB parameters from our ab-initio calculations and find that the third neighbor jeff = 1/2 hopping channel (t3) in these materials dominates over the first (t1) and second (t2) neighbor hopping channels because of cancellations of orbital overlaps within the t2g manifold in the first neighbor hopping channel. Using these estimated TB parameters, we compute edge spectra in zig-zag, armchair and Klein edge configurations. We find that these compounds have topological edge states with linear dispersion at momentum points kx = 0 and π points in the one-dimensional (1D) Brillouin zone only with the zig-zag boundary conditions provided that (1) t3 > t1/2 and (2) time-reversal symmetry (TRS) is preserved.

First principles band structure results and edge spectra — We first perform local density approximation (LDA) calculations with SOC and Hubbard U to analyze the electronic band structure of RuCl3 and A2IrO3[30]. These compounds have crystal structures with C2/m space group symmetry, consist-
of each layer, in which edge-sharing metal (M) - anion (X) top of each other \[16, 17, 20, 21, 31\]. Fig. 1(a) is a schematic expressed as \(j\) level can be mapped to a minimal single-orbital model of the showed that, due to SOC, electronic structures near the Fermi differences compared to the X- and Y-bonds. The nearest neighboring (2NN) Z-bonds in Fig. 1(a) develop small try is slightly broken and the first-nearest (NN) and second-nearest neighboring (2NN) Z-bonds in Fig. 1(a) develop small monoclinic distortion, the threefold rotation symme-

\[
\text{RuCl}_3 \quad -4.3 \quad -8.0 \quad -44.4 \quad (8.8, 8.6, 8.3) \\
\text{Na}_2\text{IrO}_3 \quad -6.3 \quad -5.3 \quad -38.2 \quad (11.4, 11.4, -2.5) \\
\text{Li}_2\text{IrO}_3 \quad -14.8 \quad -7.2 \quad -31.3 \quad (20.8, 20.8, 0.8)
\]

TABLE I. (Units in meV) Hopping integrals \(t_1, t_2\) and \(t_3\) in Eq. 1, tabulated for three different compounds. Small bond dependence for hopping integrals is taken into account by averaging over the three inequivalent bonds. For 2NN, only \(t^{\alpha}\) is shown and \(t^{X,Y}\) are presented in the Supplementary Materials.

Fig. 1(a) is a schematic of each layer, in which edge-sharing metal (M) - anion (X) octahedra MX\(6\) form a honeycomb lattice of M ions. Because of small monoclinic distortion, the threefold rotation symmetry is slightly broken and the first-nearest (NN) and second-nearest neighboring (2NN) Z-bonds in Fig. 1(a) develop small differences compared to the X- and Y-bonds.

Previous theoretical studies on the above compounds showed that, due to SOC, electronic structures near the Fermi level can be mapped to a minimal single-orbital model of the \(J_{\text{eff}} = 1/2\) states on the M-sites\[14, 25, 32\], which can be expressed as

\[
H = \left( t_1 \sum_{\langle i,j \rangle} C^\dagger_i C_j + t_2 \sum_{\langle \langle i,j \rangle \rangle} C^\dagger_{i} C_{j} + t_3 \sum_{\langle \langle \langle i,j \rangle \rangle \rangle} C^\dagger_{i} C_{j} \right) + i \sum_{\langle i,j \rangle \in \gamma} \nu_{ij} C^\dagger_{i} T^{\gamma} \cdot \sigma C_{j} + \text{h.c.} \tag{1}
\]

where \(C_i\) is the annihilation operator for the \(J_{\text{eff}} = 1/2\) pseudospinor \((\epsilon_{i\uparrow}, \epsilon_{i\downarrow})\) at site \(i\), \(\gamma\) denotes the bond type as in Fig. 1(a) and \(T^{\gamma} = (t_{2g}^{1/2}, t_{2g}^{2/2}, t_{2g}^{3})\) are bond-dependent 2NN hopping terms with \(\sigma = (\sigma_x, \sigma_y, \sigma_z)\) being Pauli matrices for the pseudospin subspace. The phases \(\nu_{ij} = \pm 1\) depend on A (+1) or B (-1) sublattice hopping. Projecting the \(J_{\text{eff}} = 1/2\) dominated subspace onto the minimal model using Wannier orbitals for each compound in our ab-inito calculations yields the model parameters tabulated in Table I. A common feature is the predominating third-nearest neighbor (3NN) \(t_3\) and 2NN \(T^{\gamma}\) terms compared to the NN \(t_1\) and 2NN \(t_2\). The small value of \(t_1\), which seems counterintuitive considering the size of NN \(t_{2g}\) hopping terms (~100 meV)\[14, 33, 34\], is the result of cancellation within the NN \(t_{2g}\) hopping channels originating from the edge-sharing geometry and the form of the \(J_{\text{eff}} = 1/2\) orbitals. Fig. 1(b) shows the two most significant channels participating in \(t_1\), the \(\delta\)- and \(\sigma\)-like overlap integrals between the neighboring \(t_{2g}\) orbitals. Since the two channels have opposite signs to each other, cancellation between them yields vanishingly small values of \(\sim 10\) meV for \(J_{\text{eff}} = 1/2\) \(t_1\) in all three compounds. On the contrary, 2NN and 3NN channels are enhanced by the presence of different channels, in addition to the \(J_{\text{eff}} = 1/2 - 3/2 - 1/2\) virtual processes which are effective beyond the NN channels (See Supplementary Materials for further details). Note that the small \(t_1\) leads to the weak NN Heisenberg term in the spin Hamiltonian as reported in Ref. 34 and 35.

Fig. 2 shows the edge spectra of RuCl\(_3\), Na\(_2\)IrO\(_3\), and Li\(_2\)IrO\(_3\) for zig-zag and armchair boundary configurations. Linearly dispersing edge states exist on the zig-zag boundary (Fig. 2(e1)) while the armchair boundary does not support any such states (Fig. 2(e2)). The degenerate points at \(k_z = 0\) and \(\pi\) on the zig-zag boundary are protected by TRS, which will be discussed in detail in the following section.

**Tight-binding model and edge states** — To understand the
TABLE II. Coefficients of the $\Gamma$ matrices in Eq. (2). We have chosen a momentum space basis such that $\mathbf{k} = k_x \mathbf{b}_1 + k_y \mathbf{b}_2$, where $\mathbf{b}_i$ are the reciprocal lattice vectors corresponding to the primitive lattice vectors $\mathbf{a}_{1,2}$ chosen as in Fig. 2(a).

| $d_0(\mathbf{k})$ | $d_1(\mathbf{k})$ | $d_2(\mathbf{k})$ | $d_3(\mathbf{k})$ | $d_4(\mathbf{k})$ | $d_5(\mathbf{k})$ | $\Gamma$ |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|--------|
| $2t_{2} \{ \cos k_x + \cos k_y + \cos(k_x - k_y) \}$ | $t_{3} \{ \cos(k_{x} - 2k_{x}) + 2\cos k_y \}$ | $-t_{3} \sin(k_y - 2k_x) - t_{1} \{ \sin(k_x - k_y) + \sin k_x \}$ | $-2(t_{2y} \sin k_x - t_{2z} \sin k_y + t_{2y} \sin(k_y - k_x))$ | $2t_{2} \{ \cos k_x - t_{2z} \sin k_y + t_{2y} \sin k_x \}$ | $-2(t_{2y} \sin k_x - t_{2z} \sin k_y + t_{2y} \sin(k_y - k_x))$ | $I_{4} \otimes \sigma_{\tau_{z}}$ |

FIG. 3. (Color online) Understanding the $t_{3}$-only limit for zig-zag and armchair boundary configurations. (a) Two big honeycombs form zig-zag (pink) and Klein (blue) boundary configurations on the zig-zag boundary of the underlying lattice. (b) Zig-zag boundary spectrum with colored edge states corresponding to the two big honeycombs. (c) The two big honeycombs both form armchair boundary configurations when the underlying lattice forms an armchair boundary. (d) No edge states are observed in the armchair spectrum.

Nature of the Hamiltonian (1) and the edge spectra, let us consider the TB Hamiltonian which takes the form $H = \sum_{\mathbf{k}} \psi_{\mathbf{k}}^{\dagger} h(\mathbf{k}) \psi_{\mathbf{k}}$:

$$h(\mathbf{k}) = d_{0}(\mathbf{k}) I_{4} + \vec{d}(\mathbf{k}) \cdot \vec{\Gamma}.$$  \hspace{1cm} (2)

We define the momentum space spinor in terms of $j_{eff} = 1/2$ operators by $\psi_{\mathbf{k}} = \{ c_{\mathbf{A}+}, c_{\mathbf{B}+}, c_{\mathbf{A}2}, c_{\mathbf{B}2} \}$. The components of the vector $\vec{d}(\mathbf{k})$ are given in Table II and $\vec{\Gamma} = (I_{2} \otimes \tau_{x}, I_{2} \otimes \tau_{y}, \sigma_{x} \otimes \tau_{z}, \sigma_{y} \otimes \tau_{z}, \sigma_{z} \otimes \tau_{z})$ is a vector of $4 \times 4$ matrices composed of sublattice $\tau$ and pseudospin $\sigma$ Pauli matrices. For the moment we ignore the identity term $d_{0}$ because it merely shifts the energy spectrum by a momentum-dependent constant and does not affect the existence of the edge states.

To comprehend the linearly dispersing edge states and their dependence on the boundary conditions, we provide a systematic analysis of the TB model below. Recall that the NN model has been widely studied[36-40] and it harbors flatband edge states on the zig-zag boundary for $|k_x| \geq 2\pi/3$, with no edge states appearing at $k_x = 0$. The current results go beyond the NN model. We note that the 3NN-only TB model involves two separated bigger honeycombs as shown in Fig. 3(a,c) with red and blue colors, and that the edge states at $k_x = 0$ may come from the Klein boundary of the blue honeycomb in Fig. 3(a). To prove this rigorously, we first investigate the $t_{3}$-only limit, then we show the effects of adding the NN $t_{1}$ term, followed by spin-dependent 2NN terms.

The $t_{3}$-only limit given by $h(\mathbf{k}) = d_{1}(\mathbf{k}) \tau_{x} + d_{3}(\mathbf{k}) \tau_{y} \equiv [d_{1}(\mathbf{k}), d_{2}(\mathbf{k}), 0]$ with $t_{1} = 0$ possesses chiral symmetry because it anticommutes with $\tau_{z}$. We follow the winding number prescription discussed in an earlier work[38] and project the Hamiltonian onto different edge configurations by cutting the lattice as in Fig. 3(a,c) and Fourier transforming along the $x$ direction to obtain a set of 1D Hamiltonians parameterized by $k_x$, denoted by $h_{k_x}(\mathbf{k}_y)$. As $k_y$ is varied from 0 to $2\pi$, $h_{k_x}(\mathbf{k}_y)$ traces a loop in $\tau$-space and if it contains the origin, the existence of degenerate edge states is guaranteed. The $t_{3}$-only model on the zig-zag boundary gives $h_{k_x}(\mathbf{k}_y) = -t_{3} \{ \cos(k_y - 2k_x) + 2\cos k_x, -\sin(k_y - 2k_x), 0 \}$, which always contains the origin thus giving flatbands for all $k_x$ in the 1D Brillouin zone, as evidenced in Fig. 3(b). For the armchair edge, $h_{k_x}(\mathbf{k}_y) = -t_{3} \{ \cos(k_y - 2k_x) + 2\cos k_x, \sin k_x, 0 \}$ which does not enclose the origin for any $k_x$ so no edge states appear as shown in Fig. 3(d). On the Klein edge, the two big honeycombs terminate as zig-zag edges so no edge states appear at $k_x = 0$. The edge spectrum of the $t_{3}$-only limit does not qualitatively change when adding $t_{1}$, except for small regions of $k_x$ so long as $t_{3} > \frac{1}{2} t_{1}$. We confirm that the corresponding winding number for all $k_x$ where the edge state exists is $\pm 1$.

We find that by adding the $d_{5}(\mathbf{k}) \sigma_{x} \tau_{z}$ term, the system is in a QSH phase with spin Chern number 2. The edge spectrum on a zig-zag boundary is shown in Fig. 4(a). When $t_{3}$ is increased so that $t_{3} = \frac{1}{2} t_{1}$, a band inversion occurs in the bulk at the M point and the model enters a non-trivial $Z_{2}$ phase. The condition that $t_{3} > \frac{1}{3} t_{1}$ is satisfied in all three compounds due to the cancellation of orbitals at the NN level discussed in the previous section. $h(\mathbf{k})$ can be block-diagonalized into pseudospin up (+) and down (−) sectors. Within each sector we project $h(\mathbf{k})$ onto the zig-zag boundary to give $h_{k_x,\pm}(\mathbf{k}_y) = [d_{1}(\mathbf{k}), d_{2}(\mathbf{k}), \pm d_{3}(\mathbf{k})]$ and find that $h_{k_x,\pm}$ wind around the origin at only $k_x = 0$, $\pi$ and a third point whose position depends on the TB parameters. We therefore expect zero-energy edge states for each pseudospin at these points in the 1D Brillouin zone. The zero-energy edge states, denoted by black dots in Fig. 4(a), are two-fold degenerate Kramers pairs.

When including the spin-flip terms $d_{3}(\mathbf{k}) \sigma_{y} \tau_{z}$ and $d_{4}(\mathbf{k}) \sigma_{y} \tau_{z}$, only the degenerate edge states at time-reversal-invariant-momentum (TRIM) $k_x = 0$ and $\pi$ remain on the zig-zag boundary. The edge spectrum of the Hamiltonian on the zig-zag boundary using ab-initio TB parameters for RuCl$_3$ is plotted in Fig. 4(b). The corresponding 1D Hamiltonian is given by $h_{k_x}(\mathbf{k}_y) = [d_{1}(\mathbf{k}), d_{2}(\mathbf{k}), d_{3}(\mathbf{k}) \sigma_{x} + d_{4}(\mathbf{k}) \sigma_{y} +$
The degeneracy at TRIM points is protected by TRS. We used a careful analysis of further neighbor exchange interactions if TRS is present. The current study also implies that effective spin models for these materials with strong SOC requires a careful analysis of further neighbor exchange interactions and their dependence on lattice optimization. For example, a recent study found that the 3NN Heisenberg term is important in determining the magnetic ground state in Na$_2$IrO$_3$ and RuCl$_3$[34].

Since there are two Kramers pairs of edge states, the effects of disorder and interactions deserve some discussion. The stability of the $Z_2$ topological insulator is limited to single-particle scattering processes because the backscattering between Kramers partners is prevented by TRS[3]. This was pointed out by Xu and Moore in Ref. [41]. There it was shown that multi-particle scattering processes may localize the edge modes in the case of an odd number of Kramers pairs. On the other hand, for two Kramers pairs of edge states such as in the current case, it was found that the edge states are stable if repulsive interactions between Kramers partners are stronger than interactions between Kramers pairs. We note that the two Kramers pairs in these materials are separated by large momentum, thus we expect that the intrapair interactions are stronger than interpair interactions.

Our result frames a recent ARPES measurement on Na$_2$IrO$_3$ which reported linearly dispersing surface states at the $\overline{\Gamma}$ point[26]. We speculate that the surface states centered at the $\overline{\Gamma}$ observed in Ref. 26 originate from the zig-zag boundary of Na$_{1/2}$IrO$_3$ single layers exposed on the surface of Na$_2$IrO$_3$ above the magnetic transition temperature. We propose the fabrication of single layers of these honeycomb systems to search for the first realization of 1D topological edge states in 2D honeycomb transition metal materials with strong SOC. In particular, $\alpha$-RuCl$_3$ is the best candidate among them since the interlayer coupling is extremely weak, possibly allowing for the creation of single layers by the graphene Scotch tape method.

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