Monolayer of the 5d transition metal trichloride OsCl₃: A playground for two-dimensional magnetism, room-temperature quantum anomalous Hall effect, and topological phase transitions

Xian-Lei Sheng¹,² and Branislav K. Nikolić¹,*
¹Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716-2570, USA
²Department of Applied Physics, Beihang University, Beijing 100191, China

(Received 9 October 2016; revised manuscript received 20 February 2017; published 3 May 2017)

Based on density functional theory (DFT) calculations, we predict that a monolayer of OsCl₃ (which is a layered material whose interlayer coupling is weaker than in graphite) possesses a quantum anomalous Hall (QAH) insulating phase generated by the combination of honeycomb lattice of osmium atoms, their strong spin-orbit coupling (SOC), and ferromagnetic ground state with in-plane easy axis. The band gap opened by SOC is $E_g \approx 67$ meV (or $\approx 191$ meV if the easy axis can be tilted out of the plane by an external electric field), and the estimated Curie temperature of such an anisotropic planar rotator ferromagnet is $T_C \lesssim 350$ K. The Chern number $C = -1$, generated by the manifold of Os $t_{2g}$ bands crossing the Fermi energy, signifies the presence of a single chiral edge state in nanoribbons of finite width, where we further show that edge states are spatially narrower for zigzag than armchair edges and investigate edge-state transport in the presence of vacancies at Os sites. Since 5d electrons of Os exhibit both strong SOC and moderate correlation effects, we employ DFT+$U$ calculations to show how increasing on-site Coulomb repulsion $U$ gradually reduces $E_g$ while maintaining $C = -1$ for $0 < U < U_c$, leads to a metallic phase with $E_g = 0$ at $U_c$, and opens a gap of topologically trivial Mott insulating phase with $C = 0$ for $U > U_c$.

DOI: 10.1103/PhysRevB.95.201402

Introduction. The quantum anomalous Hall (QAH) insulator is a recently discovered [1,2] topological electronic phase where strong spin-orbit coupling (SOC) and ferromagnetic ordering conspire to generate band gap $E_g$ in the bulk of a two-dimensional (2D) electron system, as well as conducting (i.e., gapless) chiral edge states at its boundaries. Its topologically nontrivial band structure is characterized by a nonzero Chern number $C$ counting the number edge states whose energy-momentum dispersion threads the gap of finite-width wires, while their wave functions have finite spatial extent around the wire edges. Experimental confirmation of QAH insulators is based on the observation of quantized Hall conductance in the absence of any external magnetic field [2].

Unlike a closely related quantum Hall (QH) insulator, where chiral edge states allow a spin-unpolarized electron to propagate in only one direction, or quantum spin Hall (QSH) insulator, where helical edge states appear in pairs with different chirality and spin polarization [3], the edge states of QAH insulator allow only one spin species to flow unidirectionally. Thus, the edge state transport in nanowires made of QAH insulator is robust against both magnetic and nonmagnetic disorder, which makes them superior to edge states of QSH insulator where electrons can be backscattered by disorder (such as magnetic impurities) breaking the time-reversal symmetry. The QAH insulator is also superior in potential applications to QH insulator since the latter requires large external magnetic field and low temperatures.

However, QAH insulators have been observed thus far only at very low temperatures $\lesssim 100$ mK [1,2], thereby ignoring intense theoretical and computational searches [4] for systems whose both $E_g/k_B$ and the Curie temperature $T_C$ are higher than the room temperature. Finding such materials, or heterostructures of different materials [5,6], would open new avenues for nanoelectronic and spintronic devices with ultralow dissipation where edge states act as “chiral interconnects” whose resistance is independent of the length of the wire [1].

The seminal work on the Haldane model [7], where quantized Hall conductance is found for an electronic system defined on the honeycomb lattice with SOC in the absence of an external magnetic field, has inspired search for realistic materials exhibiting QSH or QAH insulating states where the honeycomb lattice structure is often the first ingredient [8,9]. For example, graphene with enhanced intrinsic SOC [3,10,11] can be transformed into QSH insulator and then converted into QAH insulator by adding exchange magnetic field (via impurities, doping, or proximity effect) in order to suppress one of the two helical edge states of the QSH insulator. Systems predicted to realize this concept include

$^*$bnikolic@udel.edu

FIG. 1. (a) Top and (b) side view of the crystalline structure of monolayer OsCl₃ where a sheet of transition metal Os atoms is sandwiched between two sheets of Cl atoms. The Os atoms form a honeycomb lattice, and each Os atom is located at an octahedral site between the Cl atom sheets. (c) First Brillouin zone of monolayer OsCl₃ with the high symmetry points indicated.
that monolayer OsCl$_3$ possesses an intrinsic QAH insulating temperature [1]. In this Rapid Communication, we predict insulating materials which are, however, quite rare at room intrinsic sensibility of $E_g$ transition metal oxide heterostructures [9,15]. However, all additional level splitting of the surface functionalization [8]. Finally, a honeycomb lattice introducing exchange interaction via magnetic adatoms [14] or intrinsic SOC, could be converted into QAH insulator by silicene, germanene, and stanene, which already possess strong other [16,17]).

Thus, recent efforts [4,18] have also focused on the possibility of intrinsic QAH insulator realized in ferromagnetic insulating materials which are, however, quite rare at room temperature [1]. In this Rapid Communication, we predict that monolayer OsCl$_3$ possesses an intrinsic QAH insulating phase characterized by: an energy gap $E_g \approx 67$ meV, opened only when SOC is turned on in our density functional theory (DFT) calculations; a Chern number $|C| = 1$, which (i.e., the corresponding number of edge states per boundary) can be increased by tuning the Fermi energy $E_F$; and an estimated Curie temperature $T_C \lesssim 350$ K. We emphasize that $E_g$ of OsCl$_3$ is much larger than recently predicted $E_g \approx 20$ meV of the intrinsic QAH insulator in monolayer kagome lattice graphene decorated with heavy 5$d$ transition metal adatoms [12] or heterostructures like graphene/antiferromagnet [5] and graphene/ferromagnetic-insulator [6,13]. The proximity SOC in graphene can be further enhanced by inserting a monolayer of transition metal dichalcogenides (TMD) into such heterostructures [11]. Similarly, honeycomb lattices of silicene, germanene, and stanene, which already possess strong magnetic adatoms none has been realized thus far (in part, due to difficulties in keeping adatoms sufficiently apart from each other [16,17]).

The layered nature of several transition metal trichlorides $M$Cl$_3$—where $M = Ti, V, Cr, Fe, Mo, Ru, Rh, Ir$—has been explored long before [19] the present focus on the layered materials like graphene, TMDs, and Bi-based three-dimensional topological insulators. The single layer of $M$Cl$_3$ consists of a Cl-$M$-Cl sandwich where a sheet of $M$ atoms is sandwiched between two sheets of Cl atoms, with edge-sharing OsCl$_6$ octahedra forming a honeycomb lattice, as illustrated in Fig. 1. The weakness of van der Waals forces holding layers of $M$Cl$_3$ together is illustrated in Table I where we compare their interlayer binding energy, defined as $E_b = (E_{\text{monolayer}} - E_{\text{crystal}}) / n (|\vec{a} \times \vec{b}|)^{-1}$, with that of popular layered materials like graphite and Bi$_2$Se$_3$. Here $E_{\text{monolayer}}$ is the total energy of monolayer unit cell, $E_{\text{crystal}}$ is the total energy of a three-dimensional crystal unit cell, $n$ is the number of layers in a three-dimensional crystal unit cell, and $|\vec{a} \times \vec{b}|$ is the area of the unit cell.

Monolayer OsCl$_3$ is largely unexplored among transition metal trichlorides where, e.g., monolayer RuCl$_3$ as SO-

Cs$_2$Mn$_2$F$_{12}$ [18] or $E_g \approx 25$ meV for LaX ($X$ =Br, Cl, and I) compounds [4]. In addition, accurate estimates of $T_C$ for such low-dimensional magnets, which take into account magnetocrystalline anisotropy (MCA) induced by strong SOC, are lacking.

http://…/201402(R) (2017)

TABLE I. Interlayer binding energy $E_b$ (in meV/Å$^2$) of selected transition metal trichlorides MCl$_3$. For comparison, $E_b$ of typical layered materials like graphite (note that computed value for graphite is comparable to experimental estimate $E_b = 23.3 \pm 1.9$ meV/Å$^2$ [20]) or Bi$_2$Se$_3$ is also included.

<table>
<thead>
<tr>
<th>$M$Cl$_3$</th>
<th>VCl$_3$</th>
<th>FeCl$_3$</th>
<th>RuCl$_3$</th>
<th>graphite</th>
<th>Bi$_2$Se$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_b$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OsCl$_3$</td>
<td>14.4</td>
<td>17.3</td>
<td>18.1</td>
<td>19.3</td>
<td>26.4</td>
</tr>
</tbody>
</table>

FIG. 2. Electronic band structure of monolayer OsCl$_3$ with Os spins in the: (a) in-plane FM$^*$ configuration shown in Fig. 4(a) and (c) out-of-plane FM$^*$ configuration (see also Table II). The band structure is computed with GGA (solid lines) or GGA+SOC (dashed lines). (b) Density of states corresponding to panel (a). (d) The phase diagram of OsCl$_3$ with Os spins in the FM$^*$ configuration calculated by GGA+SOC+$U$.
The effect of the on-site Coulomb repulsion of Os. For monolayer OsCl$_3$, Fig. 2(d) unveils a transition from phases in Ir and Os oxides, we also use DFT calculations for semi-infinite sheets where Fig. 3 confirms that ferromagnetic configuration FM as spins in FM configuration in (a) rotate within the $xy$ plane.

The topologically nontrivial band structure of OsCl$_3$ is brought about by the combination of honeycomb lattice formed by Os ions, strong SOC of heavy transition metal osmium, and ferromagnetic ordering of their magnetic moments. Our predictions are based on DFT calculations of electronic structure of an infinite sheet of OsCl$_3$ in Fig. 2, as well as on tight-binding calculations for semi-infinite sheets where Fig. 3 confirms the presence of one chiral state per edge associated with $C ≈ 1$. While theoretical and computational searches for QAH insulators have largely been focused on finding materials or heterostructures with nonzero (and as large as possible) $C$ characterizing their bulk band structure, we show that understanding of a variety of possible spin orderings on the honeycomb lattice [24] (see Fig. 4) or spatial and transport properties of chiral edge states (see Sec. IV in the SM [27]) preserves their own attention.

In addition, motivated by the fact that comparable energy scales associated with SOC and Coulomb interaction were found to lead to a variety of competing emergent quantum phases [21,30–32] in Ir and Os oxides, we also use DFT combined with Hubbard $U$ correction (DFT+U) [33] to investigate the effect of the on-site Coulomb repulsion of 5$d$ electrons of Os. For monolayer OsCl$_3$, Fig. 2(d) unveils a transition from QAH insulator with $C ≠ 0$ to Mott insulator with $C = 0$ as $U$ is increased toward some critical value $U_\text{c}$. In Fig. 2(d) we find $U_\text{c} \simeq 0.35$ eV, but $U_\text{c}$ increases with decreasing lattice spacing $a$ (reaching $U_\text{c} \simeq 0.6$ eV at the likely experimental value for $a$ [26], as discussed in Sec. II in the SM [27]).

**Magnetic orderings of Os atoms.** Surprisingly, despite a requirement for large SOC to open sizable $E_g$, studies of candidate QAH insulators often naively assume that their magnetization is perpendicular to the plane of a 2D electron system [1], thereby neglecting SOC-generated MCA. The MCA is fundamental property of any magnet which selects energetically favorable magnetization directions (such as easy-axis, easy-plane or easy-cone) and determines stability of that direction [12]. Moreover, MCA is crucial to evade the Mermin-Wagner theorem according to which $T_C ≈ 0$ for isotropic Heisenberg ferromagnet with finite-range interactions.

Since in-plane magnetization or magnetic field cannot by itself induce quantized Hall conductance, some studies of potential QAH insulators have proposed to apply external electric field to change MCA energy so that easy-axis tilts out of the plane. However, even *in-plane magnetization* can generate QAH effect in crystals with preserved inversion symmetry but broken out-of-plane mirror reflection symmetry (i.e., $z \rightarrow −z$) [35], as satisfied by the lattice shown in Fig. 1. Another important issue is that ferromagnetic ordering, where all spins point in or out of the plane of OsCl$_3$, might not be the ground state of magnetic moments residing on the sites of the honeycomb lattice [24].

Therefore, prior to analyzing electronic band structure of monolayer OsCl$_3$, we first investigate total energy $E_{\text{tot}}$ of four possible in-plane magnetic configurations shown in Fig. 4(a), as well as of the ferromagnetic one where spins point out of the plane (FM$^+$) and the paramagnetic (PM) one. Table II shows that ferromagnetic configuration FM$, whose spins point along the x axis, is the ground state (even at finite $U$, see Fig. S3 in the SM [27]). Additionally, Fig. 4(b) shows that energy $E_{\text{tot}}$ increases if Os spins try to move away from the x axis as the easy axis. This means that monolayer OsCl$_3$ is an anisotropic 2D XY ferromagnet [36], or, more precisely, “planar rotator” ferromagnet as a special case of 2D XY model where $S_z ≈ 0$. Its spin-spin interactions are described by an effective Hamiltonian $H_I = −\sum_{\langle ij \rangle} J_{ij}(S_i^+ S_j^- + S_i^- S_j^+) − \sum_j D(S_j^z)^2$, where $(S_i^x, S_i^y)$ is the spin operator in quantum [36] or unit vector [37] in the classical version of the model, $(i, j)$ denotes the summation over nearest neighbors, and $D$ quantifies anisotropy ($D \rightarrow \infty$ suppresses fluctuations in the $y$ component of the spins, thereby leading to the Ising model). The 2D system described by $H_I$ with sufficiently large $D/J$ undergoes transition from FM$^+$ to FM at the Curie temperature which we estimate as $T_C < 750$ K for $D/J = 0.4$ according to Table II and Fig. 4(b) [for $D/J \lesssim 10^{-7}$], one expects transition from FM$^+$ to Berezinskii-Kosterlitz-Thouless (BKT) phase with topological order, and then to PM at $T_{\text{DKT}}$. The interplane exchange coupling described by an additional Hamiltonian, $H_L = −\sum_{\langle ij \rangle} J_L(S_i^x S_j^x + S_i^y S_j^y)$, would further increase $T_C$ of OsCl$_3$ multilayers, whose spin Hamiltonian is given by $H_I + H_L$ with $J_L/J_H ≈ 0.1$ estimated from DFT calculations for OsCl$_3$ bilayer.

**TABLE II.** The total energy $E_{\text{tot}}$ per unit cell (in meV, relative to $E_{\text{tot}}$ of FM$^+$ ground state), as well as spin $⟨S⟩$ and orbital $⟨O⟩$ moments (in $\mu_B$), for several magnetic configurations of Os atoms (first four of which are illustrated in Fig. 4) calculated by GGA+SOC method. Paramagnetic state has $E_{\text{tot}} = 47.36$ meV and $⟨S⟩ = ⟨O⟩ = 0$.

<table>
<thead>
<tr>
<th>FM$^+$</th>
<th>NAFM$^+$</th>
<th>SAFM$^+$</th>
<th>ZAFM$^+$</th>
<th>FM$^+$</th>
<th>FM$^+$</th>
<th>NAFM$^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{\text{tot}}$</td>
<td>0.0</td>
<td>39.60</td>
<td>47.57</td>
<td>16.63</td>
<td>27.42</td>
<td>0.51</td>
</tr>
<tr>
<td>$⟨S⟩$</td>
<td>0.57</td>
<td>0.13</td>
<td>0.10</td>
<td>0.41</td>
<td>0.30</td>
<td>0.57</td>
</tr>
<tr>
<td>$⟨O⟩$</td>
<td>0.30</td>
<td>0.24</td>
<td>0.90</td>
<td>0.33</td>
<td>0.12</td>
<td>0.30</td>
</tr>
</tbody>
</table>
Topological bulk band structure. Figures 2(a) and 2(c) show bulk band structure of monolayers OsCl$_3$ (assuming $U = 0$) in the FM$^+$ and FM$^-$ configuration of Os spins, respectively, computed using DFT implemented in the VASP package [39]. The electron core interactions are described by the projector augmented wave (PAW) method [40], and we use Perdew-Burke-Ernzerhof parametrization of generalized gradient approximation (GGA) for the exchange-correlation functional. The cutoff energy for the plane wave basis set is 500 eV for all calculations. The k-point mesh $10 \times 10 \times 1$ is used for the Brillouin zone integration in the self-consistency cycle. For the density of states (DOS) and $E_{\text{tot}}$ in Table II we use finer meshes of $25 \times 25 \times 1$ and $32 \times 32 \times 1$ points, respectively. We fully optimize the atomic coordinates until Hellmann-Feynman forces on each ion are less than 0.01 eV/Å, which yields the lattice constant $a_0 = 5.99$ Å (note that a recent x-ray study finds $a = 5.87$ Å [26]).

When SOC is switched off, Fig. 2(a) shows that monolayer OsCl$_3$ exhibits nonzero exchange splitting between spin-up and spin-down bands, but remains metallic. Switching SOC on opens a band gap $E_g \approx 67$ meV around $E_F$, as highlighted by the DOS in Fig. 2(b). Furthermore, this gap is topologically nontrivial as indicated by the nonzero Chern number $C = -1$ (note that we find $C = 3$ for VCl$_3$ and $C = -1$ for RuCl$_3$, but $C = 0$ for FeCl$_3$, CoCl$_2$, and IrCl$_4$). Figure 2(c) shows that $E_g \approx 191$ meV would be even larger for FM$^-$ configuration of spins, but this requires us to apply a very large out-of-plane electric field due to the large difference between $E_{\text{tot}}$ of FM$^+$ and FM$^-$ configurations in Table II. The Chern number $C = 1$ is obtained for FM$^+$ configuration of spins in Fig. 2(c), which also shows how $C$ could increase [28] by tuning $U_F$ (such as by gate voltage [41]) into different gaps of the bulk band structure in Fig. 2(c).

We compute $C$ from the Kubo formula [42] using the $k$-space Hamiltonian obtained by Fourier transforming the real-space tight-binding Hamiltonian (TBH) in the basis of maximally localized Wannier functions [43]. Since low-energy physics is mainly dominated by 5$d$ orbitals of transition-metal atoms, we use all five $d$ orbitals centered on each Os atom, while $p$ orbitals on Cl atoms are neglected. The on-site potential and hopping through the fourth-nearest neighbor are calculated directly from the GGA+SOC Hamiltonian generated by DFT calculations. Moreover, the DOS in Fig. 2(b) shows that the crystal field splits $d$ orbitals into $t_{2g}$ and $e_g$, where only three $t_{2g}$ orbitals contribute to the DOS around $E_F$ (whereas the contribution of $p$ orbitals of Cl is much smaller).

Energy-momentum dispersion of chiral edge state. We also use Wannier TBH to obtain the local DOS at the zigzag or armchair edge of a sheet of OsCl$_3$ which is infinite in the $x$ direction and semi-infinite in the $y$ direction. The local DOS, computed from the retarded Green function as $-\text{Im} \hat{G}^r(E; k_x, j)/\pi$ [44], is plotted in Fig. 3. It confirms the presence of one chiral edge state, whose $E_k$ dispersion penetrates through the $E_g$ gap, in accord with $|C| = 1$ and bulk-boundary correspondence [45]. The spatial and quantum transport properties of chiral edge states in OsCl$_3$ nanoribbons with different edge termination, including the role of the kink [45] in $E_k$ dispersion in Fig. 3(c) on the transmission function in the presence of disorder, are discussed in the SM [27].

Correlation-driven topological quantum phase transition. Although the accurate value of $U$ is not known for OsCl$_3$, we expect moderate correlation effects due to spatially extended 5$d$ orbitals of Os. Therefore, similarly to recent studies of electronic phases in other Os compounds [32] we vary $U$ from 0 to 1.5 eV in Fig. 2(d), with effective $U = U^* - J$ chosen in Dudarev parametrization of DFT+$U$ where the exchange parameter $J$ is not treated separately. This reveals that the QAH insulating phase persists up to some critical value $U_c \approx 0.35$ eV (for its dependence on the lattice constant see the SM [27]), at which $E_g \to 0$ due to electron correlations. Further increase of $U > U_c$ opens a gap of SO-coupled Mott insulating phase which is topologically trivial with $C = 0$. Thus, transition from QAH insulator to Mott insulator is an example of a continuous (due to gap closing at $U_c$) topological (due to change of topological number $C$) quantum phase transition [46].

Conclusions. Using DFT and DFT+$U$, as well as MCA energy and quantum transport, calculations we predict that monolayer OsCl$_3$ offers an unforeseen playground to examine electronic phases governed by the interplay between SOC, low-dimensional magnetism stabilized by large anisotropy due to strong SOC, and correlations of 5$d$ electrons. Upon increasing the on-site Coulomb repulsion, monolayer OsCl$_3$ undergoes a quantum phase transition from QAH insulator to correlated metal and finally to topologically trivial SO-coupled Mott insulator. Interestingly, a large gap $E_g = 67$ meV with nonzero Chern number $C = -1$ is observed even though its spontaneous magnetization is along the in-plane easy axis, which is possible when the mirror reflection symmetry is broken [35]. We note that Ref. [31] has proposed a heuristic phase diagram in which the QAH insulating phase borders [28] a trivial Mott insulator. Thus, our Fig. 2(d) can be viewed as a prescription for realizing such phase diagrams by using realistic materials whose 2D nature makes it possible to manipulate its charge density and $E_F$ by the gate electrode (as demonstrated very recently in correlated monolayer TMDs [41]). The correlations of 5$d$ electrons characterized by spatially extended wave functions can be captured more accurately with DFT+dynamical mean field theory [47], which we relegate to future studies.

Acknowledgements. We thank Jiadong Zang for valuable insights. This work was supported by NSF Grant No. ECCS 1509094. The supercomputing time was provided by XSEDE, which is supported by NSF Grant No. ACI-1053575.
MONOLAYER OF THE 5d TRANSITION METAL ... PHYSICAL REVIEW B 95, 201402(R) (2017)

[21] See Supplemental Material (which includes Refs. [48–53]) at http://link.aps.org/supplemental/10.1103/PhysRevB.95.201402 for the analysis of: phonon band structure of OsCl3 in the presence and absence of SOC (Sec. I), lattice spacing dependence of the critical value of Hubbard U_c (Sec. II), E_{int} of different magnetic configurations as a function of increasing Hubbard U (Sec. III), and spatial and transport properties of chiral edge states in zigzag and armchair nanoribbons made of OsCl3 in the QAH insulator phase (Sec. IV).

In this Supplemental Material we provide additional results for: phonon band structure of OsCl$_3$ in the presence and absence of SOC which allows to examine stability of monolayer OsCl$_3$ (Sec. I); lattice spacing dependence of the critical value of Hubbard $U_c$ (Sec. II); total energy $E_{\text{tot}}$ of different magnetic configurations as a function of increasing Hubbard $U$ (Sec. III); and spatial and transport properties of chiral edge states in zigzag and armchair nanoribbons made of OsCl$_3$ in the QAH insulator phase with $U = 0$ (Sec. IV).

I. PHONON BAND STRUCTURE IN THE PRESENCE AND ABSENCE OF SPIN-ORBIT COUPLING

Figure S1 shows phonon band structure of monolayer OsCl$_3$. Since the effect of spin-orbit coupling (SOC) on phonons can be profound on surfaces and in thin films of materials containing heavy elements, Fig. S1 plots phonon frequency-momentum dispersion in the presence of SOC and FM$^x$ magnetic ordering from Fig. 4(a) of the main text. Comparing this result (dashed line in Fig. S1) with phonon band structure of monolayer OsCl$_3$ where SOC is artificially turned off (solid line in Fig. S1), indeed confirms the importance of including SOC in phonon calculations. In both cases, we find no imaginary frequencies, which ensures that the lattice structure of monolayer OsCl$_3$ is at least metastable. We note that in the paramagnetic monolayer OsCl$_3$ without SOC, whose energy in Table II of the main text is much higher than the ground state with FM$^x$ ordering and SOC, we do find some imaginary phonon frequencies.

The phonon calculations were performed on the primitive unit cell using density functional perturbation theory as implemented within the VASP and PHONOPY\textsuperscript{[1]} packages. The details of VASP calculations are the same as in the main text, but with the cutoff energy for the plane wave basis set chosen as 400 eV and a mesh of $5 \times 5 \times 1$ used for $k$-point sampling. The electronic self-consistent convergence parameter was set at $1 \times 10^{-6}$ eV.

![Phonon Band Structure](image)

Fig. S1: The phonon band structure of monolayer OsCl$_3$ with both SOC and FM$^x$ magnetic ordering from Fig. 4(a) in the main text included (dashed line). For comparison, we also show the phonon band structure in the artificial case of FM$^x$ magnetic ordering where SOC is turned off (solid line).

*Electronic address: bnikolic@udel.edu
II. DEPENDENCE OF THE CRITICAL VALUE OF HUBBARD $U_c$ ON THE LATTICE CONSTANT

Applications of DFT+U methodology to different materials have revealed sensitivity of Hubbard $U$ on the lattice spacing of the unit cell, such as in the case of iron where a marked increase of $U$ occurs as the lattice constant is squeezed below its experimental value \cite{2}. In Fig. S2, we show the dependence of the critical value $U_c$, at which topological quantum phase transition takes place in Fig. 2(d) in the main text, on the lattice spacing. We used $a_0 = 5.99$ Å in the main text, as obtained from DFT optimization of the atomic coordinates, but the experimental lattice constant appears to be slightly smaller $a = 5.87$ Å \cite{3} which would increase $U_c$ from $\simeq 0.35$ eV found in Fig. 2(d) in the main text to $U_c \simeq 0.6$ eV at $a/a_0 \approx 0.98$ in Fig. S2. Although there are no extensive experimental studies of OsCl$_3$ to confirm its lattice constant obtained in Ref. \cite{3}, this estimate seems compatible with experimental lattice constants as one moves from FeCl$_3$, where $a = 6.13$ Å, to RuCl$_3$, where $a = 5.9783$ Å \cite{4}, and eventually to OsCl$_3$ with the heaviest transition metal atom among the three isoelectronic ones.

III. DEPENDENCE OF THE TOTAL ENERGY OF DIFFERENT MAGNETIC CONFIGURATIONS ON HUBBARD $U$

All calculations of the total energy $E_{tot}$ for different magnetic configurations in Fig. 4 and Table II of the main text are performed at $U = 0$. Figure S3 shows $E_{tot}$ as a function of increasing $U$, which demonstrates that FM$^x$ configuration in Fig. 4(a) remain the ground state even at finite $U$. 

Fig. S2: Dependence of the critical value of Hubbard $U_c$ from Fig. 2(d) in the main text on the lattice constant $a$ of monolayer OsCl$_3$ in units of $a_0 = 5.99$ Å used in the main text.

Fig. S3: The total energy $E_{tot}$ per unit cell, relative to $E_{tot}$ of FM$^x$ ground state, for several magnetic configurations of Os atoms in Fig. 4 of the main text as a function of Hubbard $U$. 

Using Wannier tight-binding Hamiltonian discussed in the main text, we compute the real-space retarded Green function [7]

\[ \hat{G}^r(E; i, j) = [E - \hat{H} - \hat{\Sigma}_L - \hat{\Sigma}_R]^{-1} \]  

(1)

of the central region of OsCl₃ nanoribbons attached to the left (L) and right (R) semi-infinite leads of the same width \( W \). The leads generate self-energies \( \hat{\Sigma}_{L,R} \) determining escape rates of electrons from the central region into the macroscopic reservoirs kept at electrochemical potential \( \mu_{L,R} \). The active region is either perfectly clean or host two vacancies (one on each edge) as defects obtained by cutting out all bonds adjacent to a chosen Os site. For example, in Dirac materials like graphene and Bi₃Se₃ vacancies drastically affect the local DOS, by introducing sharp peaks of quasilocalized character which can be detected by scanning tunneling microscopy [8], and represent infinitely strong scatterers which cannot be handled by any perturbative analysis [9].

The subband structure of clean zigzag and armchair nanoribbons is shown in Figs. S4(a) and S4(e), respectively. Using the retarded Green function and the level broadening function \( \hat{\Gamma}_{L,R} = -2\text{Im}[\hat{\Sigma}_{L,R}] \), we compute the transmission function [7]

\[ T(E) = \text{Tr}[\hat{\Gamma}_R \hat{G}^r \hat{G}_L \hat{G}^r] ] \]  

(2)

at vanishing applied bias voltage \( \mu_L - \mu_R \to 0 \), which is plotted in Figs. S4(b) and S4(f). It exhibits quantized steps in the clean case, which are destroyed outside of the bulk gap region in Figs. S4(a) and S4(e) when vacancies are introduced. However, \( T(E) \) in Fig. S4(f) has a dip even within the topologically protected bulk gap due to wide spatial extension of edge states in the armchair case, so that hybridization of edge states from opposite edges opens
a minigap at the crossing point in Fig. S4(e). The local DOS, $-\text{Im} \hat{G}^r(E; i, i)/\pi$, shown for zigzag in Fig. S4(c) and armchair nanoribbons in Fig. S4(g) confirms that edge states in the former case are narrower than in the latter case, as observed previously [5, 6] for edge states of QSH insulators emerging from different types of graphene nanoribbons.

The zigzag edge changes the Hamiltonian near the boundary which then introduces a kink in the dispersion of edge state in Figs. 3(c) of the main text and S4(a), as also found for the Haldane model defined on the honeycomb lattice with zigzag edge [10]. For some values of energy $E$ within the bulk band gap, such dispersion intersect with $E$ at $N_R = 2$ points with positive velocity and $N_L = 1$ points with negative velocity. This leads to $T(E) = 3$ for $E$ within a portion of the bulk band gap in Fig. S4(b). Nevertheless, only the difference $N_R - N_L = |C|$ is topologically protected according to bulk-boundary correspondence [10], leading to $T(E) \geq 1$ across the whole bulk band gap even in the presence of vacancies in Fig. S4(b).